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ABSTRACT (cont)

The present study extend previous work to include electron radiation-induced changes in monoisopropyl biphenyl (MIPB)-impregnated polypropylene film as well as the effects of neutron/gamma radiation on dry polypropylene films. Effects that were quite similar were induced by both electron and neutron radiation on the properties of interest of the polypropylene films. Impregnation of the film with MIPB had a mitigatory effect on the degradation of the properties. This report also contains the results of a simultaneous electrical and thermal agirg study of a capacitor-grade polypropylene film. The data obtained in this study was fitted to models that will enable realistic prediction of lifetimes under operating conditions.

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RESEARCH OBJECTIVES

To establish a theoretical and experimental base in order to study and understand multifactor stress aging of insulating and dielectric materials.

ABSTRACT

Insulating and dielectric materials used in space-based applications encounter a multitude of stresses that includes electrical, thermal and radiation fields. These stresses act singly or simultaneously to degrade the materials and to ultimately cause failure. Thus, the reliability and life of a great many space-power and other electrical components are critically dependent on the performance of the insulating and dielectric systems that they incorporate. To predict the performance or life of such systems to any degree of certainty requires a matrix of multiple stress aging data which can be used to formulate lifetime models. The present study extends the work reported in the First Annual Report to include electron radiation-induced changes in monoisopropyl biphenyl(MIPB)-impregnated polypropylene film (Chapter 2) as well as the effects of neutron/gamma radiation on dry polypropylene films (Chapter 3). Effects that were quite similar were induced by both electron and neutron radiation on the properties of interest of the polypropylene films. Impregnation of the film with MIPB had a mitigatory effect on the degradation of the properties.This report also contains the results of a simultaneous electrical and thermal aging study of a capacitor-grade polypropylene film (Chapter 4). The data obtained in this study was fitted to models that will enable realistic prediction of lifetimes under operating conditions.

CHAPTER 1

INTRODUCTION

As space power requirements move into the multi kilovolt/amp range[1], electrical insulation becomes the critical issue that has to be addressed in order to realize targeted goals. Higher energy densities and greater reliabilities that are important in land-based applications become the two most important considerations in space-based systems because of the prohibitive launch and repair/maintenance costs. Therefore the main thrusts of research in this area have been and continue to be the development of new materials, dielectric systems and capacitive geometries to increase the amount of energy that can be stored per unit weight or volume, and the investigation of the mechanisms of failure of dielectric and insulating systems in order that realistic models for predicting the lifetimes of such systems can be postulated.

Dielectrics and insulators undergo long-term failure usually under the aging action of electrical and thermal stresses. In addition, dielectrics operating in space or in nuclear reactor environments are subjected to bombardment by ionizing radiations of various kinds. Also, the smaller volume of the nuclear reactors in spacecraft and the lower efficiency of heat rejection in space contribute to higher thermal stresses[2]. Therefore, in most

cases of future generations of spacecraft, the insulating material employed will be required to operate under multiple stresses of electrical, thermal and radiation (E/T/R) fields. A single-point failure in insulation may very well prove catastrophic for the whole mission.

Although there have been a number of studies dealing with the aging of insulating materials under electrical and/or thermal stresses, they have, for the most part, dealt with high voltage cables and have not employed thin polymeric film dielectrics or high electric fields commonly found in capacitor applications. The effects of ionizing radiation on polymeric materials have been documented to some degree, although the radiation-induced changes in the dielectric properties and the breakdown characteristics have not been investigated and accounted for. Also, simultaneous multifactor aging studies under electrical, thermal and radiation (E/T/R) stresses are practically non-existent.

The work reported here is an effort to fill in some of the gaps that exist in order to move towards a comprehensive E/T/R aging study of high voltage capacitor dielectric films. Chapter 2 is an extension and continuation of the previously reported work on the effects of electron radiation on polypropylene films, in the First Annual Report and elsewhere[3-5]. Specifically, the consequences of monoisopropyl biphenyl (MIPB) impregnation on the

radiation-induced changes were studied. In Chapter 3, the effects on the electrical and chemical characteristics of polypropylene films aged by neutron and gamma irradiation in a nuclear reactor up to 10 hours are reported. The results of the lifetime studies of polypropylene films under simultaneous electrical and thermal stresses are reported in Chapter 4. In accordance with the films' use in high voltage and pulsed power capacitors, electric fields employed were high compared to those encountered in high voltage cables. The data obtained were fitted to existing models that describe simultaneous electrical and thermal aging. New research directions, as outlined in Chapter 5, comprise of systematic multifactor stress studies, extended to include radiation and the development of semi-empirical models to reliably predict the lifetimes of dielectric or insulating systems under such stresses.

The results from Chapters 2 to 4 have been written and submitted as three journal papers, one of them already published [6] and two of them still under active review[7,8].

CHAPTER 2

EFFECT OF HIGH ENERGY ELECTRON RADIATION ON MIPB-IMPREGNATED POLYPROPYLENE

2.1. INTRODUCTION

In high voltage and space power systems, there is always a demand for increasing the energy densities as well as the power levels. In addition, components with better reliability, lower losses, compactness and longer lifetimes are required. Insulating materials which are commonly employed within these systems, are often the weak links as their degradation and breakdown precipitate the failure of the equipment. The degradation is accelerated by the exposure to radiation in environments such as space and nuclear reactors. These materials, therefore, play a key role in the design and performance of these systems.

Polymers nowadays constitute the largest class of insulating materials in use because of their excellent electrical and mechanical properties.[9] Polypropylene is one such polymer which finds major application in high energy density capacitors. The polymer film is often impregnated with a dielectric fluid to improve its reliability and life by way of suppressing discharges

within voids and gas pockets. The fluid also helps in transferring away the heat that builds up during the operation of a capacitor. Monoisopropyl biphenyl (MIPB) is an aromatic hydrocarbon liquid that possesses good dielectric properties, low viscosity and high gas absorption ability.[10] In addition, this liquid is known to exhibit excellent radiation stability.[11]

To investigate the influence of radiation on a typical dielectric system used in high voltage and energy storage capacitors, MIPB-impregnated polypropylene film was exposed to high energy ionizing radiation. Electron radiation, which has been reported as influencing the performance of many space vehicles and on-board electronic components[12,13], was chosen in this work. The post-radiation effects on the electrical, mechanical, morphological and chemical properties were investigated and are reported as functions of the total absorbed dose. The electrical properties included the dielectric constant, dissipation factor, the ac (60 Hz) and dc breakdown voltages. The mechanical properties investigated comprised the Young's modulus, elongation-at-break, tensile strength, storage modulus and mechanical loss. The morphological and chemical characterizations included the determinations of crystallinity (X-ray diffractometry), surface morphology (optical and scanning electron microscopy) and chemical changes (Infrared spectroscopy and sol-gel determination).

2.2. EXPERIMENTAL

A 25.4 μm-thick biaxially-oriented polypropylene film, manufactured by Hercules, Inc., was used as the solid dielectric. The film was impregnated with MIPB, an aromatic hydrocarbon fluid. Some of the properties of interest of the film and the impregnant are listed in Tables I[14] and II[10,15], respectively.

Impregnation of the films was done as follows:Samples were first dried in a vacuum oven at 10^{-2} Torr at 50°C for 6 hours. The liquid impregnant was then bled into the oven till the samples were completely immersed in the fluid. The samples were then allowed to soak in MIPB for 16 hours at 50°C under vacuum. At the end of this period, the samples were allowed to cool slowly to room temperature and dry nitrogen was bled into the oven to bring the pressure up to atmospheric. The impregnated samples were then stored in a nitrogen atmosphere.

While the radiation procedure of the films as well as the techniques adopted in performing the electrical, mechanical, morphological and chemical measurements are given in detail elsewhere [4,5,16], it is worth enumerating the following important points of the experimental procedure:

- The films were exposed in air to 1 MeV electrons at

Table I. Properties of the polypropylene film.[14]

Dielectric constant, 24 °C, 60 Hz & 1 kHz	2.3
Dissipation factor, 24 °C, 60 Hz & 1 kHz	0.0002
Dielectric strength, 24 °C, 60 Hz (V/μm)	315
Specific gravity	0.902
Weight-percent crystallinity	56
Tensile modulus (N/mm^2)	2,420
Elongation (%)	70 - 100
Tensile strength (N/mm^2)	205
Melting point (°C)	167
Glass transition temperature (°C)	-18

Table II. Properties of MIPB.[10,15]

Classification	80% aromatic hydrocarbon
Dielectric constant, 25 °C, 60 Hz	2.8
Dielectric strength, 25 mm gap (V/μm)	2.8
Specific gravity	1.0
Viscosity, 38 °C (cs)	6.1
Solubility in water	Insoluble
Boiling point (°C)	270
Flash point (°C)	155
Pour point (°C)	-55

a rate of 10^6rads/min. Samples with total radiation doses of 10^5rads, 10^6rads, 10^7rads and 10^8rads were obtained. During each irradiation run, the samples were stacked in a tray with the excess oil being removed in order to ensure uniform exposure and to prevent the attenuation of the electron beam.

- The breakdown voltage measurements were carried out using two cylindrical brass electrodes of 2.54 cm diameter and the rate of voltage rise was 500 V/s. To prevent surface flashover, the breakdown tests were conducted in a test cell filled with transformer oil.

- Strips having the dimensions 5 cm x 6 cm were extended at a rate of 20 mm/min in carrying out the tensile tests. The dynamic mechanical properties were determined in a temperature range from 12^{o}C to 120^{o}C. These measurements were made at a frequency of 110 Hz and the rate of increase of the temperature was 3^{o}C/ min.

2.3. RESULTS AND DISCUSSION

2.3.1. Electrical Properties

Dielectric characterization of the films was carried out in terms of changes in their dielectric constant and the dissipation factor with the radiation absorbed dose at six frequencies, ranging from 50 Hz to 10 kHz.

The dielectric constant of the impregnated film did not display any dependence on the frequency. Therefore, only the data obtained at 50 Hz is represented in Figure 2.1. It can be clearly seen that the film suffered a slight reduction in its dielectric constant as the absorbed dose was increased. At 10^8rads, for example, this property had decreased by 15% of its unirradiated value. Such a reduction of the dielectric constant with increasing dose may point towards the crosslinking of the polymer.

Unlike the dielectric constant, the dissipation factor of the film exhibited a significant dependence on the frequency of measurement. The values of this property at four frequencies are plotted against the absorbed dose in Figure 2.2. While there was a significant decrease in the dissipation factor with increasing radiation dose at low frequencies, the effect is less dramatic at high frequencies. This seems to indicate that crosslinking of the

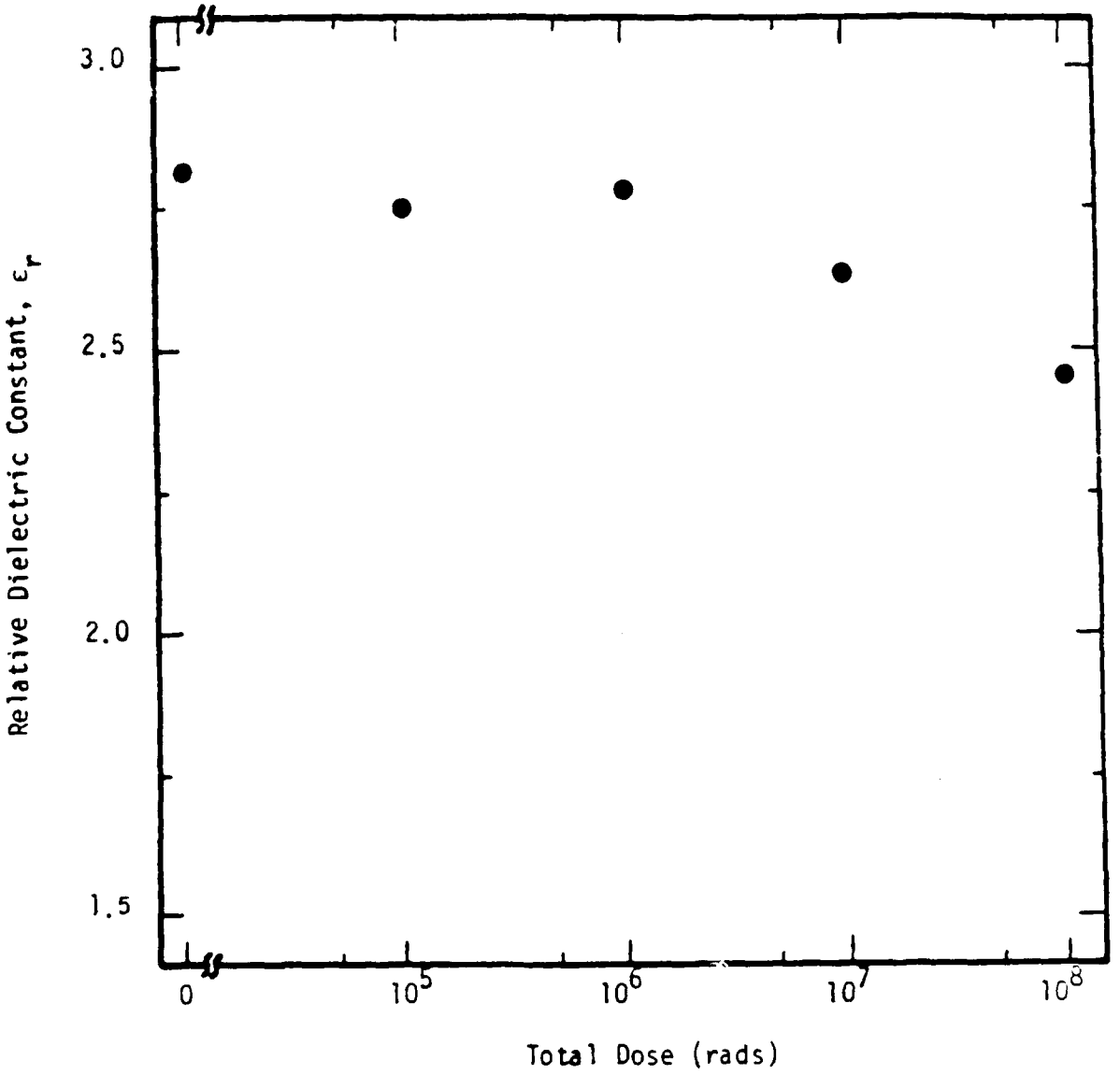

Figure 2.1. Relative dielectric constant at 50 Hz versus total radiation dose.

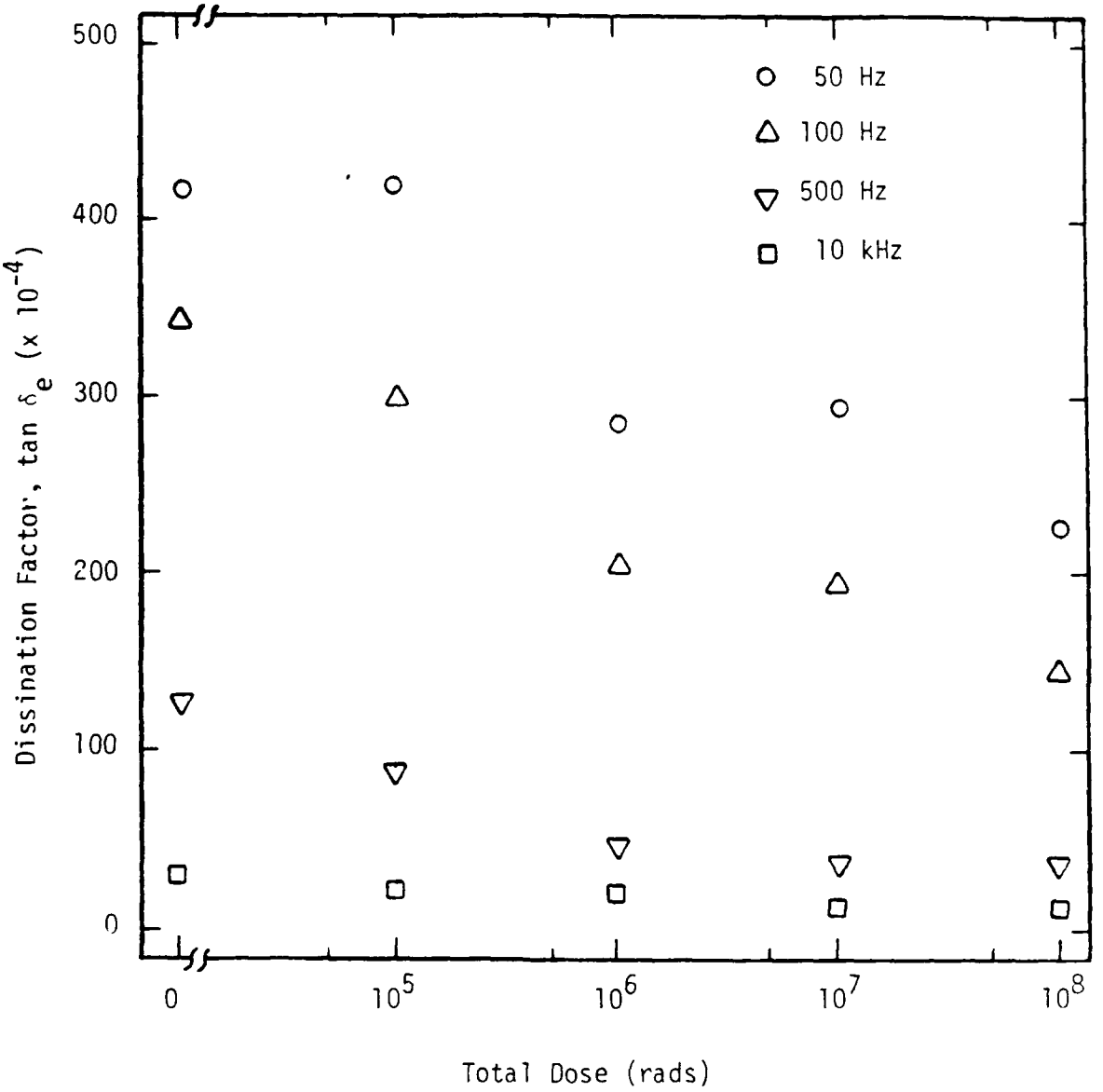

Figure 2.2. Dissipation factor versus total radiation dose.

polymer has an inhibitory effect only on the motions of the larger molecular segments which are responsible for the losses at lower frequencies. The orientational motions of the smaller molecular segments, which are presumably responsible for the losses at higher frequencies, are not affected by the large scale interlinking of the molecules.

A comparison of the ac (peak) and the dc breakdown voltages are shown in Figure 2.3. While the dc breakdown voltage dropped initially with the first exposure to radiation, it remained unaffected by further radiation. The ac breakdown voltage did not exhibit any significant changes with irradiation. Although the spread in the data was rather large, the claim can be made that impregnation of the film has lessened the impact of radiation on the breakdown behavior of the film.

2.3.2. Mechanical Properties

The variation in the Young's modulus of the impregnated film with radiation absorbed dose is shown in Figure 2.4. It is seen that the Young's modulus exhibits a gradual increase with increasing dose, reaching at 10^8rad dose level a value which was more than double of what it was before exposure to radiation. As

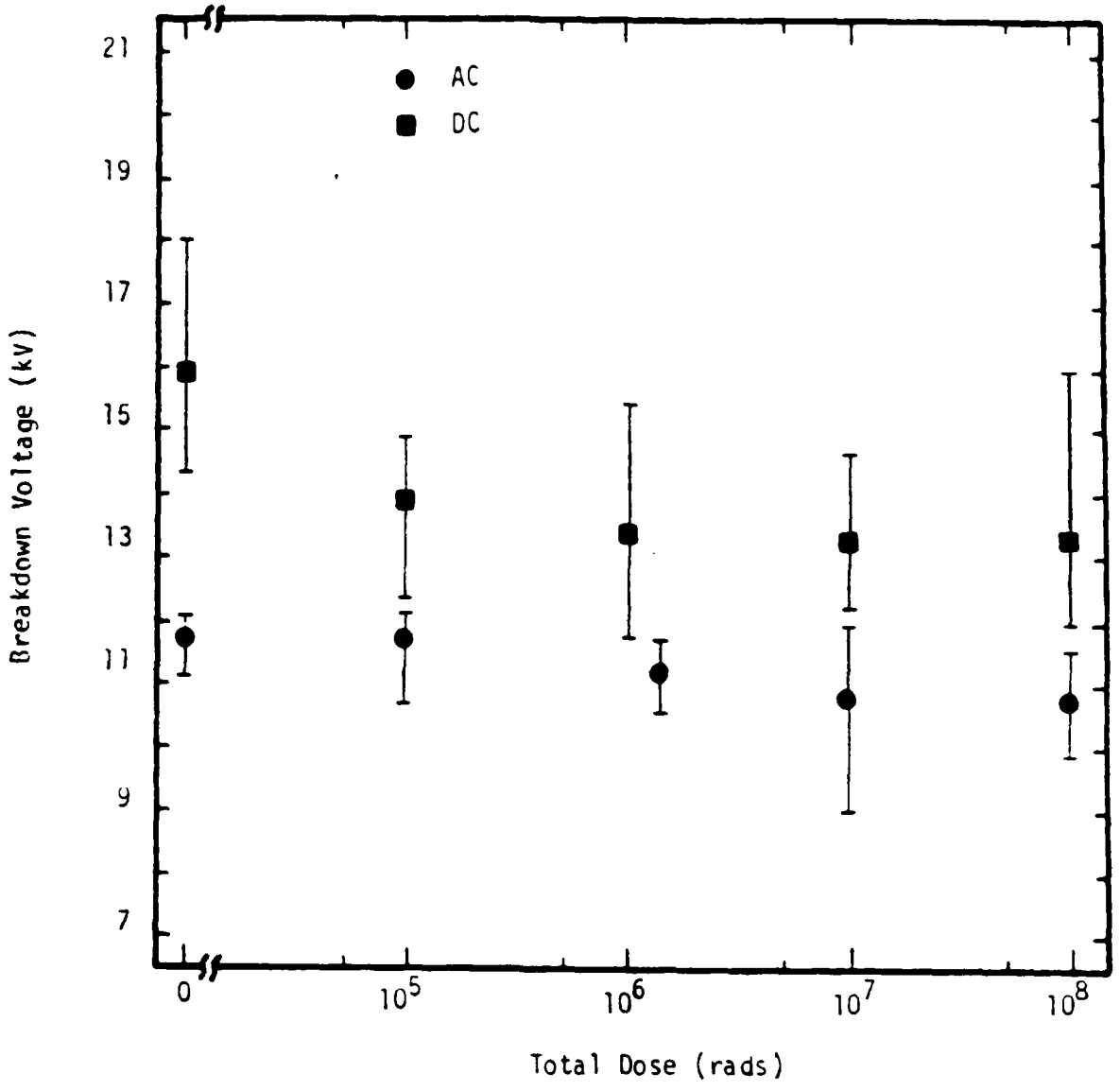

Figure 2.3. Breakdown voltage versus total radiation dose.

crosslinking is known, in general, to increase the Young's modulus of polymers [17], the data obtained support the contention that the impregnated films underwent crosslinking.

Figure 2.5 illustrates the changes in the elongation-at-break of the film as a function of the radiation absorbed dose. After showing little change with increases in the dose up to 10^6 rads, the elongation-at-break decreased with further increases in the dose. At 10^8rads, this property lost about 30% of its unirradiated value. This decrease in the material's ability to stretch at any given stress may once again be symptomatic of the crosslinked structure existing in the irradiated samples of the polypropylene film.

Unlike the Young's modulus and the elongation-at-break, the tensile strength of the film did not show any variation with radiation dose (Fig. 2.6). Hegazy et al. [18] have reported similar trends for the tensile properties of isotactic polypropylene when exposed to gamma radiation under vacuum.

The dynamic mechanical properties of impregnated films irradiated to different total doses are shown in Figures 2.7 and 2.8. The temperature range (12^oC to 120^oC) in which these measurements were made corresponds to the region between the glass transition (β) and the pre-melting relaxation ( ) of

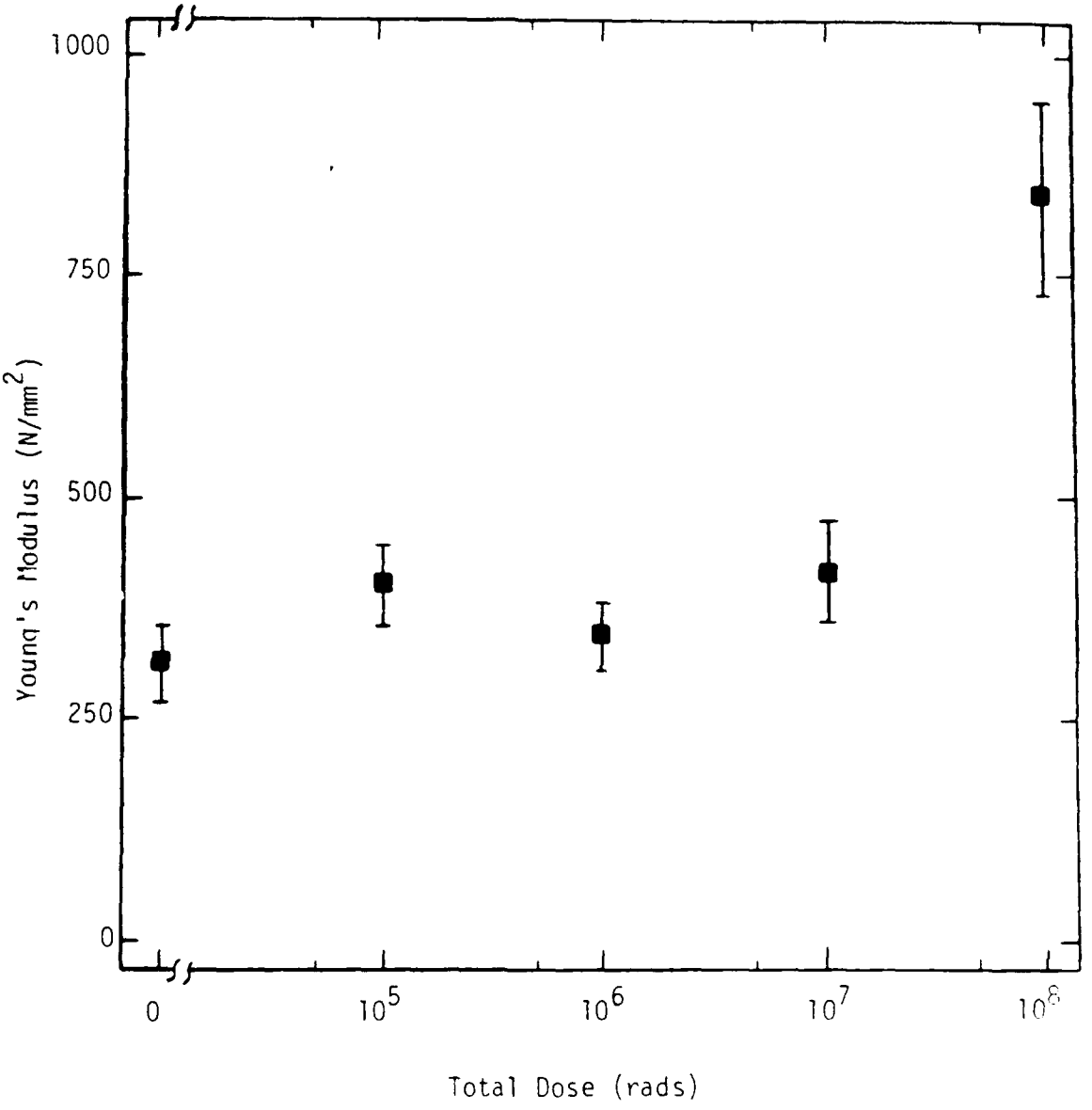

Figure 2.4. Young's modulus versus total radiation dose.

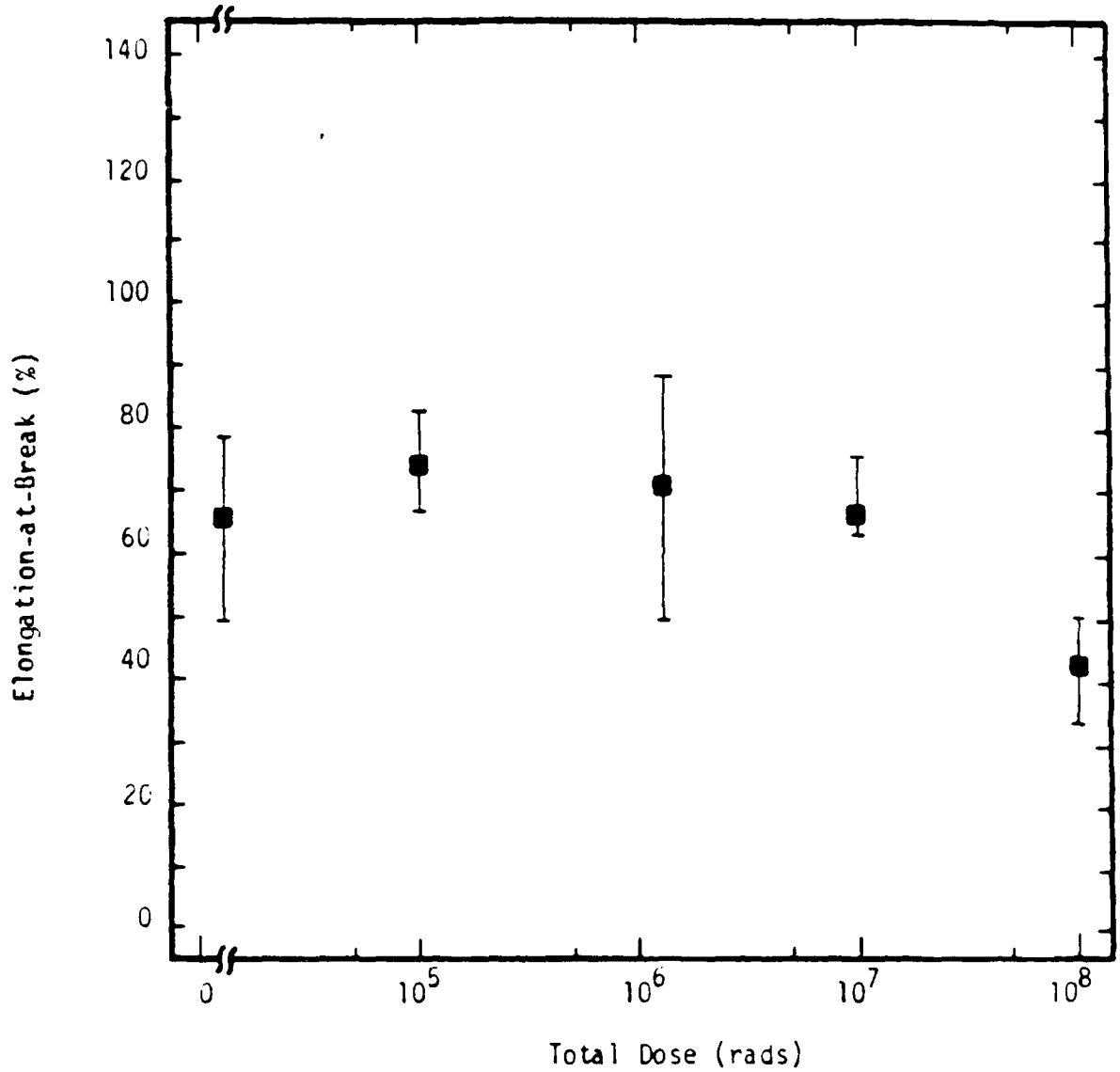

Figure 2.5. Elongation-at-break versus total radiation dose.

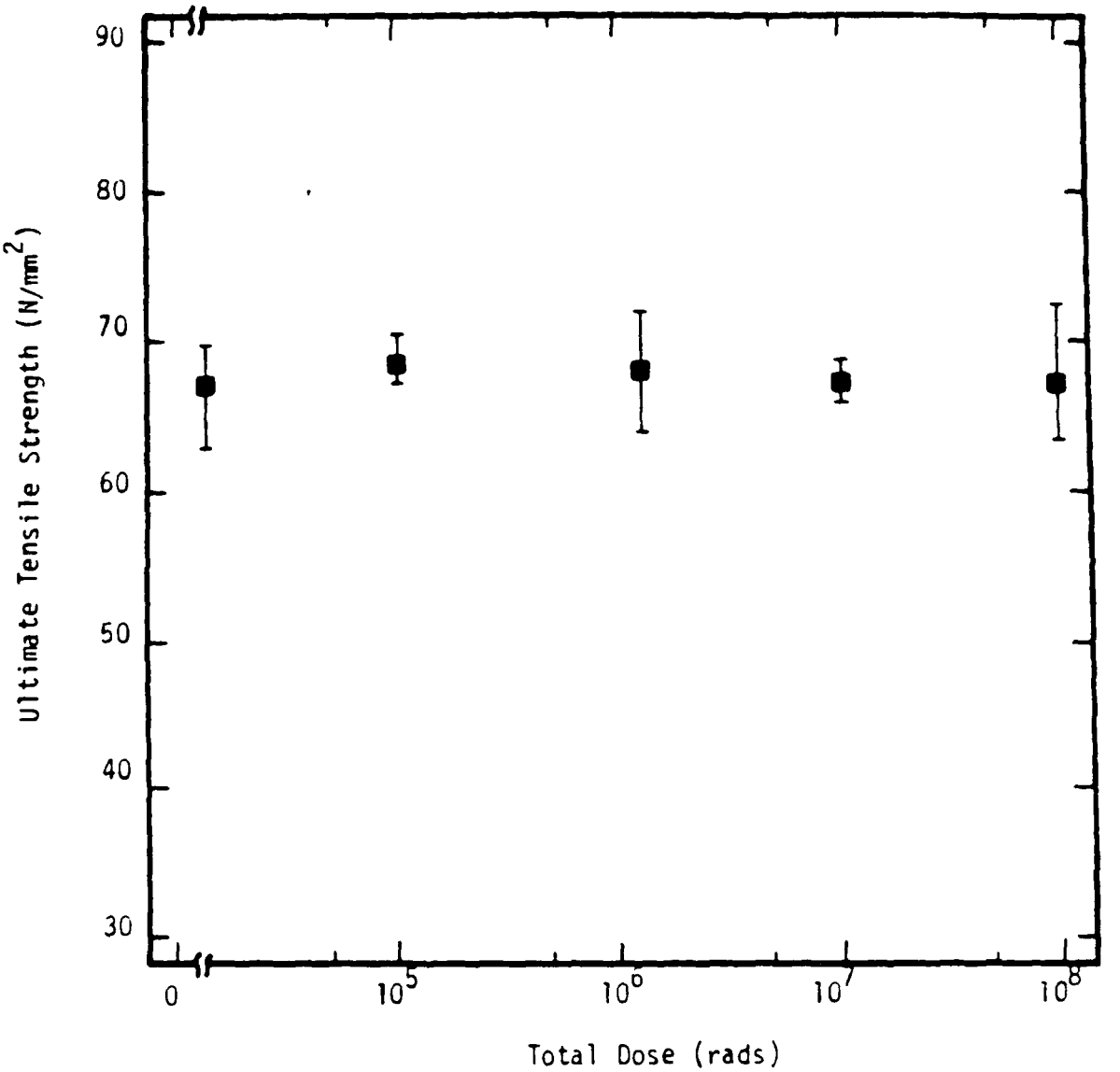

Figure 2.6. Ultimate tensile strength versus total radiation dose.

isotactic polypropylene. The data show that radiation has not caused any significant effect on either the storage modulus (Fig. 2.7) or the mechanical loss (Fig. 2.8) until a dose of 10^8rads is reached. These changes, which appear primarily above 40°C, consist in a decrease in the modulus and an increase in the loss of the 10^8rad sample. The precise nature of the α relaxation, in which region most of the changes are observed in the present study, is not unambiguously known.[19] It is possible that the destruction of the crystalline order by crosslinking is responsible for the increase in the loss or the equivalent decrease in the storage modulus of the 10^8 rad sample. Until the dynamic mechanical measurements are extended to include a wider range of temperatures (particularly below 0°C), the data pertaining to these properties would remain inconclusive.

2.3.3. Morphological and Chemical Characterization

Optical and scanning electron microscopic studies did not reveal any differences between the virgin (unirradiated) and irradiated samples. Also, no radiation-induced chemical changes, such as oxidation or the formation of unsaturated bonds, were detected through the use of the infrared spectroscopy.

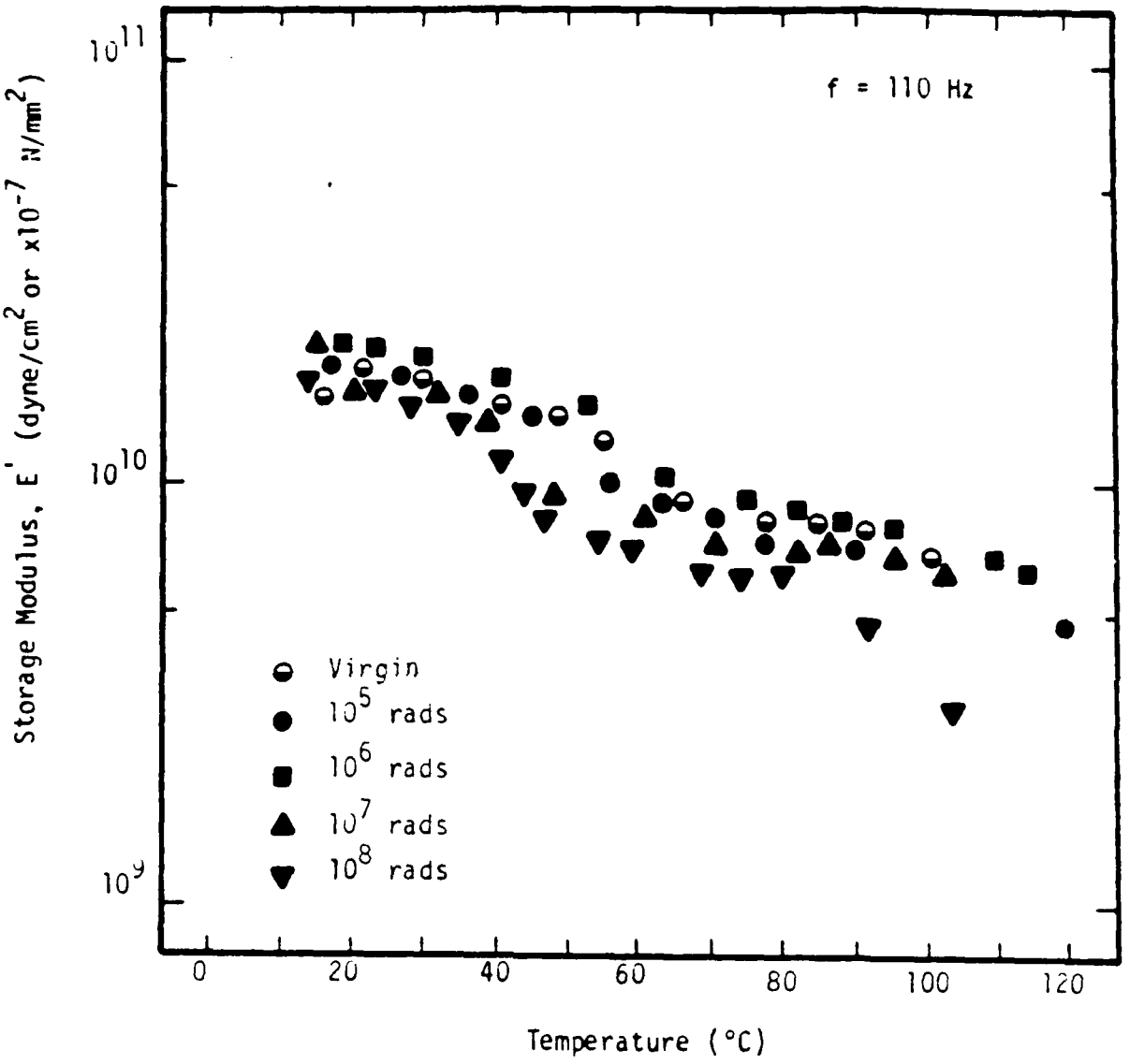

Figure 2.7. Variation in the storage modulus with temperature for different total radiation doses.

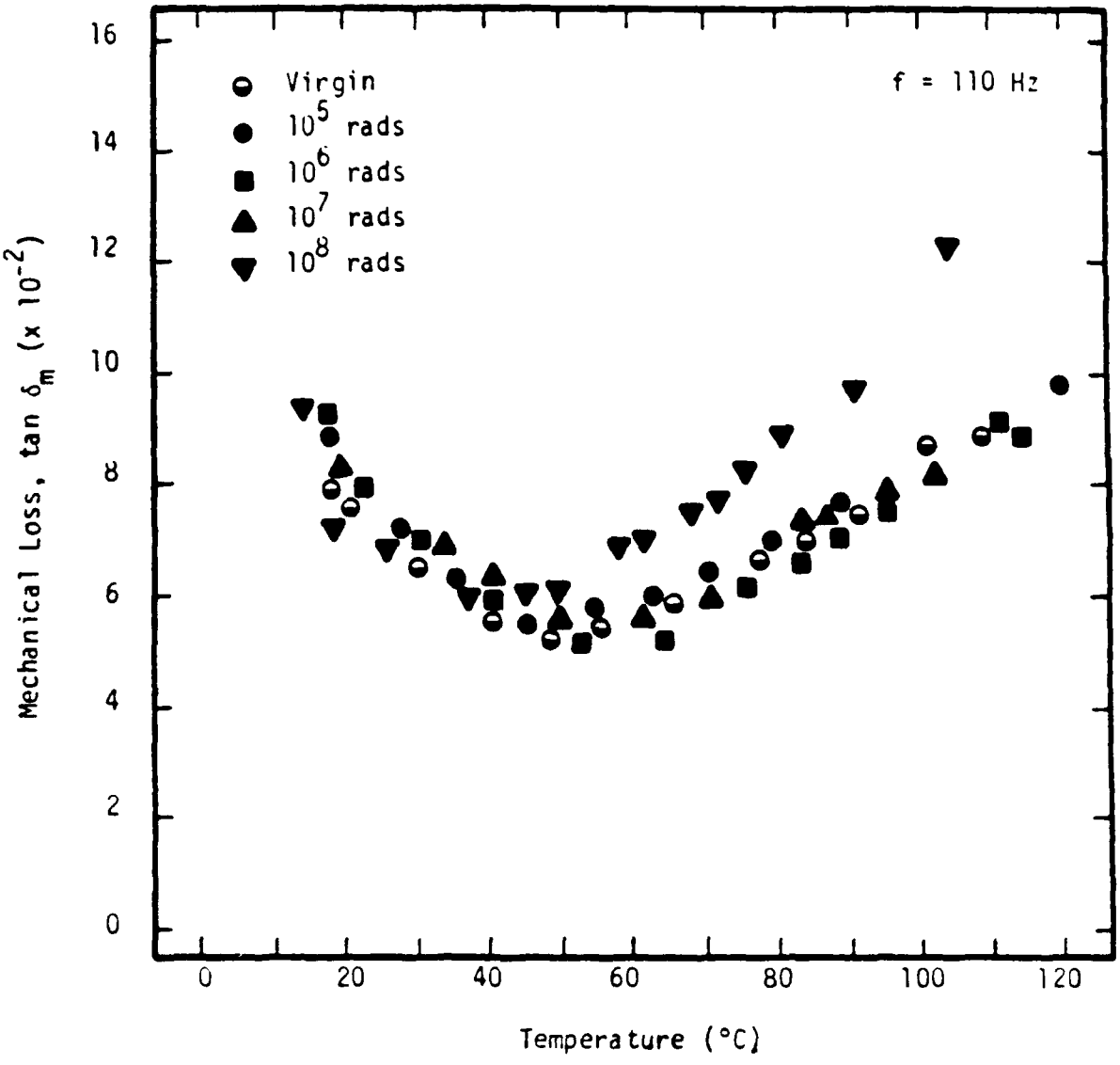

Figure 2.8. Variation in the mechanical loss tangent with temperature for different total radiation doses.

X-ray diffraction scans of the impregnated films, virgin as well as irradiated are shown in Figure 2.9. The height of the peak at $2\theta = 17^{o}$ which corresponds to the (040) planes and being the most prominent, was taken as a measure of the crystallinity of the samples. Upon exposure to radiation, the film exhibited an initial slight increase in its crystallinity with dose up to 10^{6}rads. Thereafter, the crystallinity decreased with dose to a level lower than the value at the film's unirradiated state. It is possible that the initial increase in the crystallinity was due to a molecular rearrangement facilitated by the radiative energy imparted to the polymer --- a kind of radiative annealing. At higher doses, however, crosslinking would introduce disorder in the crystalline matrix reducing the crystalline content [20].

Although no direct method for identifying crosslinking in polymers exists [20], an attempt was made by means of sol-gel determinations, to determine whether the films had indeed crosslinked upon exposure to radiation. In these experiments, samples were extracted in boiling toluene for a duration of 90 minutes. Table III shows that the gel fraction has remained essentially constant with increasing dose. The deviation at 10^{5}rads may have been due to experimental error. In contrast to those of the dry films[3-5], the present results suggest that, at the least, radiation-induced chain scission was effectively suppressed by the impregnation of the film. In fact, there is

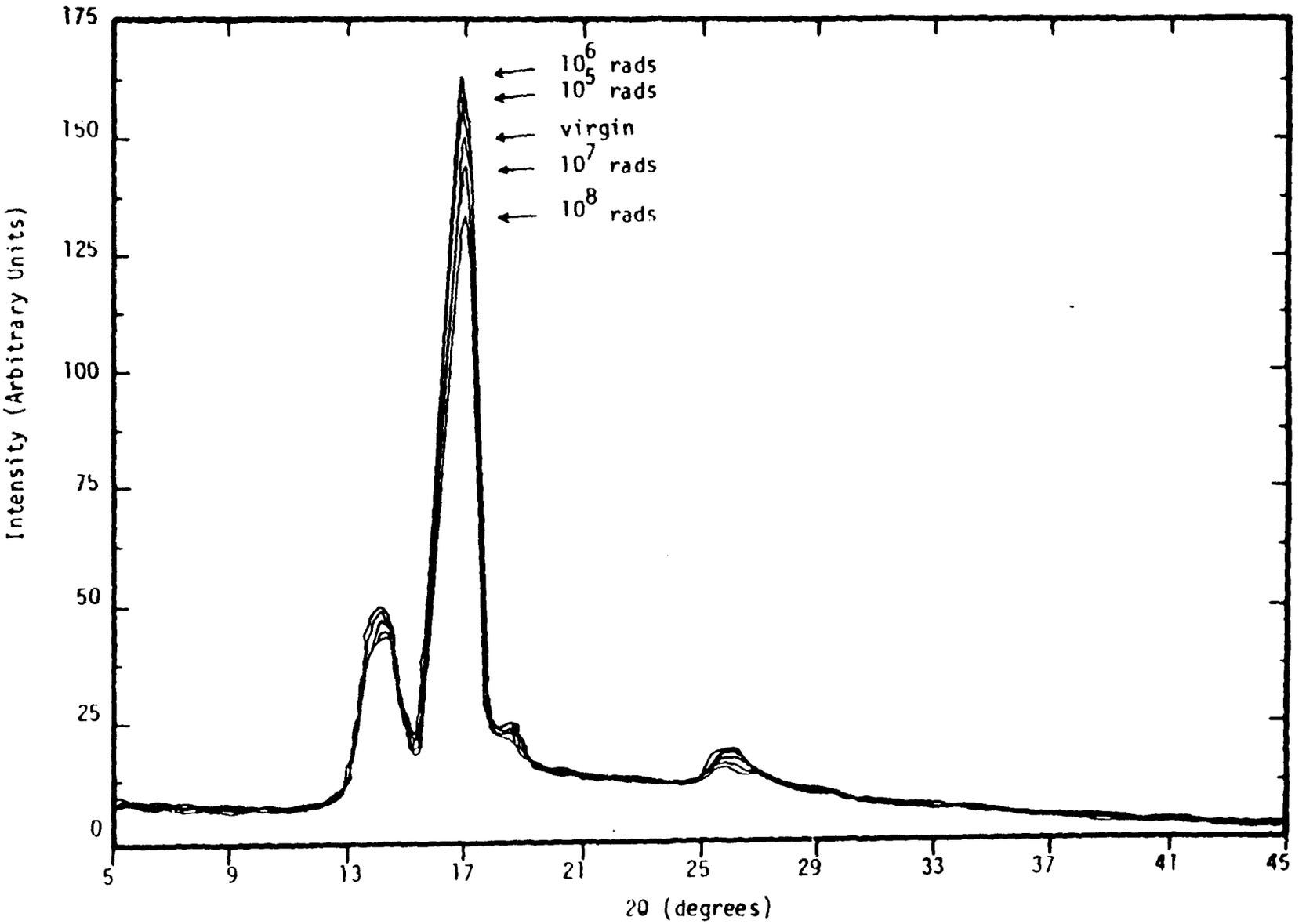

Figure 2.9. X-Ray diffraction intensity curves for virgin and radiated samples.

Table III. Gel fraction results from sol-gel determinations.

Sample	% gel
Virgin	54
10^5 rads	36
10^6 rads	52
10^7 rads	56
10^8 rads	61

some indication that crosslinking has occurred as reflected by the slight increase in the gel fraction at the high doses. It is believed that the barrier presented by the impregnating fluid to the free diffusion of oxygen during radiation has been instrumental in retarding chain scission and favoring crosslinking. These results taken together with the increase in the Young's modulus with radiation support the speculations made earlier that changes in some of the film properties are attributable to crosslinking. In addition, infrared spectroscopic studies indicated the absence of any oxidative degradation.

2.4. CONCLUSIONS

The present work indicates that polypropylene, widely employed in cables and capacitors, can be reliably used in radiation environments when they are impregnated with a suitable dielectric fluid, such as MIPB. When exposed to high energy electron radiation up to doses as high as 10^8rads, the impregnated films showed good tolerance and stability in terms of their electrical and mechanical properties. This stability to radiation comes about due to the presence of the impregnant which seemed to have encouraged crosslinking in preference to chain scission and oxidative degradation.

CHAPTER 3

EFFECTS OF FAST NEUTRON RADIATION ON POLYPROPYLENE

3.1. INTRODUCTION

As technology advances, there is a constantly growing need for systems that will perform satisfactorily in harsh environments, such as nuclear radiation.[21] Nuclear sources for electric power generation are very well established in earth-based applications. The nuclear power source is also the most promising candidate for space power needs due to the compactness of nuclear reactors as compared to other power sources. [1,22] Of first concern for materials and components are the structural metals of the core containment and the ceramic insulators of the primary control equipment and instrumentation.[23] But recently, it has also become evident that organic materials which have much lower resistance to radiation damage, are needed for service in equipment located in other areas within the containment where the radiation dose, typically 10 to 100 rad/hr., can be appreciable over the design life of the plant. [23] Also the emergency cooling equipment and monitoring controls have to operate reliably in case of interruption in the supply of coolant (Loss of Coolant Accident) or the rupture of one or more fuel elements containers. In this case the emergency equipment must con-

tinue to function even though the integrated dose could be in the range of $5x10^8$ rads.[23]

A great number of electrical components and seals are listed as being inside the containment building of a nuclear power plant. This include cables, motors, switches, relays, connectors, capacitors, transformers etc.[23] All of them in one way or another incorporate organic materials as electrical insulation. One particular device in which dielectrics play a very significant role is the capacitor. These capacitors are to be located in the containment vessels of nuclear power facilities where they are to protect the primary coolant motors from overvoltage surges.[24] The dielectric most commonly employed in capacitors is polypropylene film [10], whose behavior in the radiation environment of thermal and fast neutrons becomes, therefore, of primary importance. Unfortunately, there is very little information on the changes in dielectric properties of insulating materials, particularly polymer insulation, after irradiation in a thermal nuclear reactor. [25] This work attempts to understand the changes that can take place in polymer insulation, polypropylene in this case, in a radiation environment. The results for dielectric properties of polypropylene film such as electric strength, dielectric constant, dissipation factor and volume resistivity as a function of the total absorbed dose are given. The changes in the

electrical properties are found to be related to changes in chemical structure of the film and are described below.

3.2. EXPERIMENTAL

Capacitor grade, biaxially oriented polypropylene films (Type EK-500, manufactured by Hercules Inc.), having 25 μm thickness, were exposed to neutron and gamma radiation in a two megawatt, open-tank type reactor in the Nuclear Science and Technology Facility at Buffalo. A pneumatic system was used to position samples at the side of the reactor. Polypropylene samples were encapsulated in a sealed bag and inserted inside the "rabbit" made of molded polyethylene.

Two rolls of polypropylene films were prepared and irradiated by means of the pneumatic conveyer, one for a duration of one hour and the other for ten hours. Because of radiation damage to the rabbit, radiation runs were kept to less than five hours. Therefore, the ten hours irradiation was achieved by two five-hour cycles with two polyethylene rabbits. The total flux of neutrons over the entire range of energies was $2.6 \times 10^{12} n/cm^2 s$ accompanied by gamma radiation at the level of 10^7 Rad/hr.

Due to the low level, but still harmful for personnel residual radioactivity of the irradiated samples, at least two months decaying period was needed, until radioactivity dropped to less than 1000 cpm level, before the samples were released for tests out of the nuclear reactor site. The post-irradiation effects on the electrical and chemical properties were, therefore, studied for polypropylene films with total accumulated doses of $1x10^{16}$ and $1x10^{17}$ n/cm^2 and compared with those of the virgin film.

The electrical tests performed on irradiated and virgin films consisted of d.c. and a.c. breakdown voltages, the life test under repetitive pulse voltage stress, dielectric permittivity and dielectric losses and volume resistivity. Infrared analysis was subsequently done on the radiated films to determine the chemical changes occurring.

For a.c., d.c. and repetitive pulse voltage stresses, the power supplies used were , Hipotronics 715-5, Universal Voltronics BAM-32-1.5-B and a 25 kV Model-9 Hard-Tube Pulser, respectively. The pulsed voltage in particular, had a negative amplitude with the rate of 100 pulses per second, 2 μs pulsewidth, 0.2 μs rise time and 0.6 μs fall time. All breakdown experiments as well as the pulsed life test were carried out in a plexiglas test cell filled with transformer oil (Diala AX, Shell Oil Company),

using 2.54 cm diameter brass electrodes and a rate of voltage rise of 500 V/s in accordance with ASTM-D149 standard. All destructive tests due to their statistical nature consisted of at least seven data points, allowing therefore to show the spread in the data.

A capacitance measurement system (1621 General Radio Precision Capacitance Bridge) together with a set of concentric ring brass electrodes were used in the measurement of the dielectric permittivity and the dielectric loss tangent of the samples at room temperature. High purity silver paint was painted on the film to provide a good contact between the brass electrodes and the film.

Volume resistivity was measured using Keithley 6105 Resistivity Chamber, 2 kV d.c. power supply and Keithley 616 electrometer. The charging current was monitored with a chart recorder until a stable value was obtained from the voltage application. The chemical changes, like the formation of carbonyl groups, were monitored using infrared spectrometer Perkin-Elmer 237B IR.

The energy imparted by the ionizing radiation was converted to the energy absorbed by the polypropylene film, and is described in Appendix B. For radiation times used in the present study, the total neutron doses of $1x10^{16}$ and $1x10^{17}$ n/cm^2s were equiv-

alent to 2.9×10^7 and 2.9×10^8 rads of the energy absorbed, respectively.made of molded polyethylene.

3.3. RESULTS AND DISCUSSION

3.3.1. Electrical Properties

To characterize the dielectric properties of irradiated films, the permittivity and the dissipation factor were measured. The dielectric constant as a function of frequency for virgin and irradiated films is shown in Figure 3.1. Very slight increase of the permittivity is observed when the total absorbed dose was $1 \times 10^{16} n/cm^2 s$. With ten times higher dose, the increase is much more pronounced. Still, the approximately eight percent change is very insignificant and is believed to be caused by the slow oxidation of the film. [4] Creation of the carbonyl groups, which are highly polar, causes an increase of the dielectric constant but at the same time increases dielectric losses of the insulating film. The effect of neutron radiation on the dissipation factor of polypropylene film is shown in Figure 3. 2 . Significant increase of the dielectric losses is observed due to the oxidation of the film, which was confirmed by the infrared absorption spectra analysis, as is reported later in this chapter.

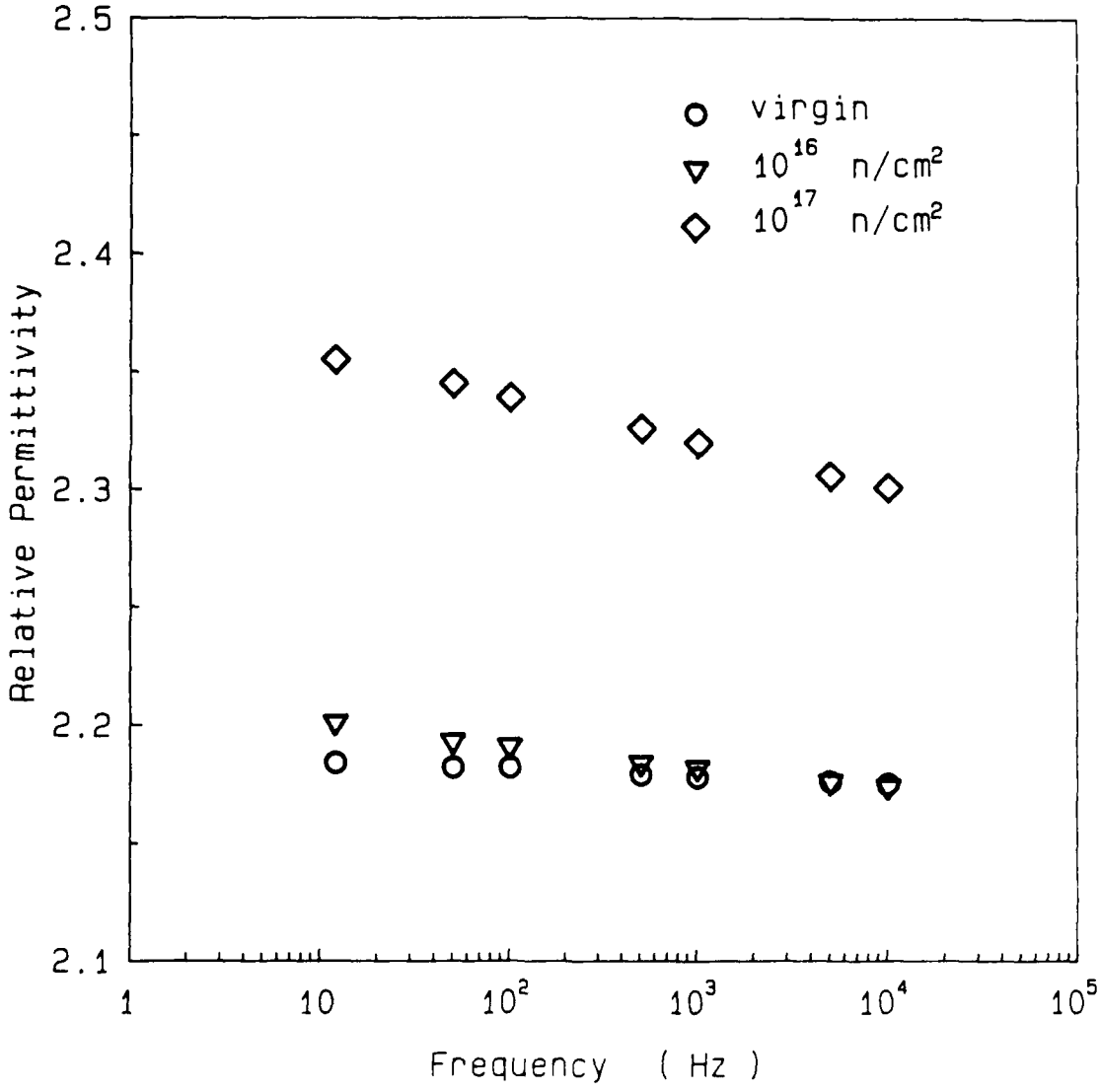

Figure 3.1. Dielectric constant versus frequency for virgin and irradiated PP films.

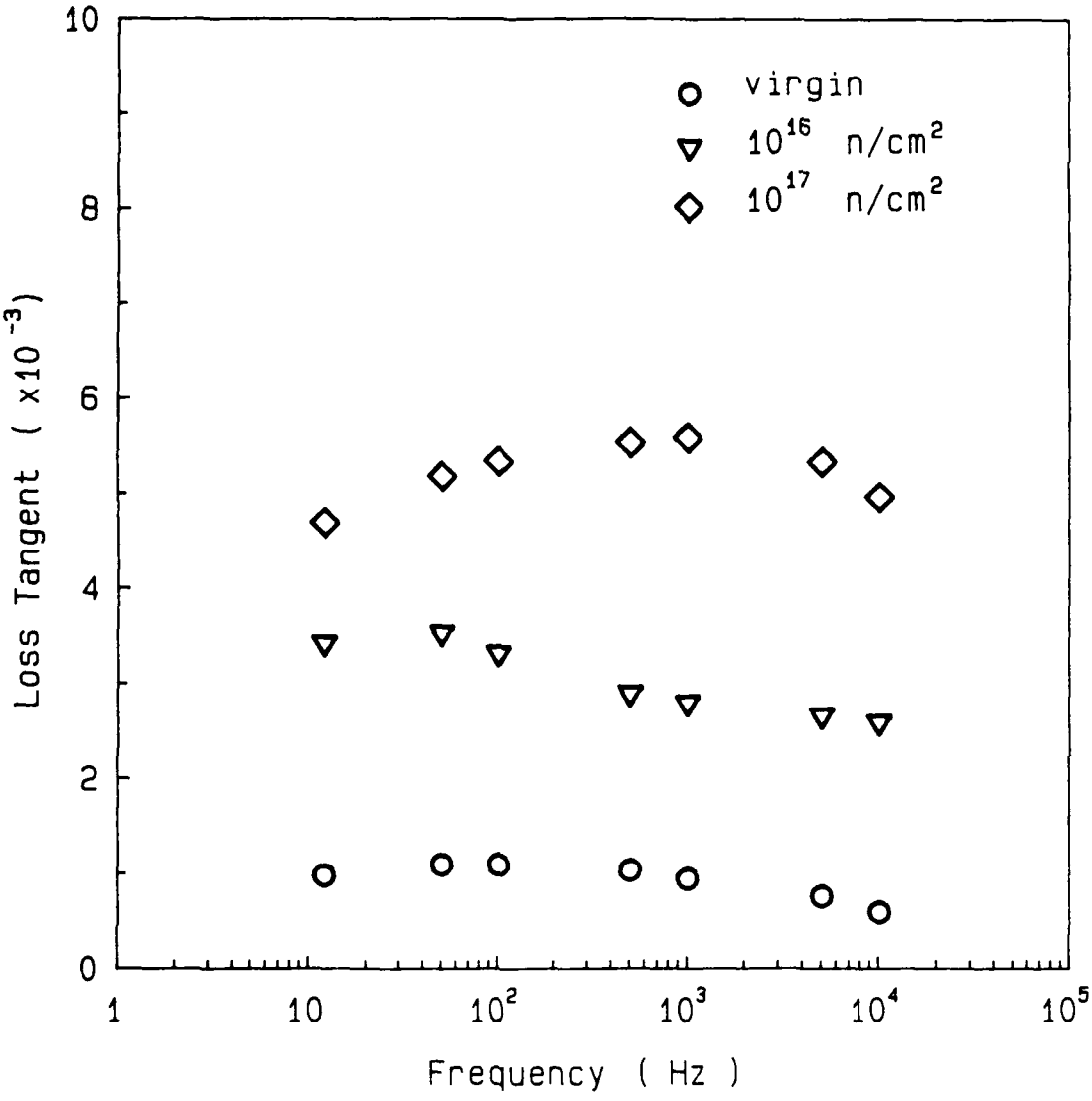

Figure 3.2. Dissipation factor versus frequency for virgin and irradiated PP films.

The ac and dc breakdown voltages, as well as the spread between the minimum and maximum values, for the 25 μm thick film are shown as a function of the total dose in Figure 3.3. The dependence of breakdown voltage on the absorbed dose of neutron radiation carries some degree of uncertainty due to the high scatter in the data, nevertheless, attempt is made to analyze the trends and mechanisms responsible for changes in the electric strength of polypropylene. For both types of voltages there was a slight, about 10 percent, increase in the breakdown strength when the total absorbed dose was $1x10^{16}$ n/cm^2s. This increase in breakdown strength can be attributed to the crosslinking of the polymer molecules, as it was also postulated by Hammoud et al[4,5] in case of electron irradiation of polypropylene. For the total absorbed dose of $1x10^{17}$ n/cm^2s, the breakdown strength decreased to values lower than that of a virgin film. Even though the crosslinking was even higher, which was evident in the high brittleness of the film, the lower breakdown voltage came about by the formation of weak spots and cracks, as could be visually observed, and the consequent degradation of the film.
In consideration of the film's application in capacitors located in nuclear radiation environment, the virgin and irradiated films were then subjected to life tests under rep-rate pulse stress to 85 percent of the pulse breakdown voltage (measured to be 14.3 kV for the 25 μm thick virgin film). The number of pulses to breakdown was measured and the results are plotted in the Figure

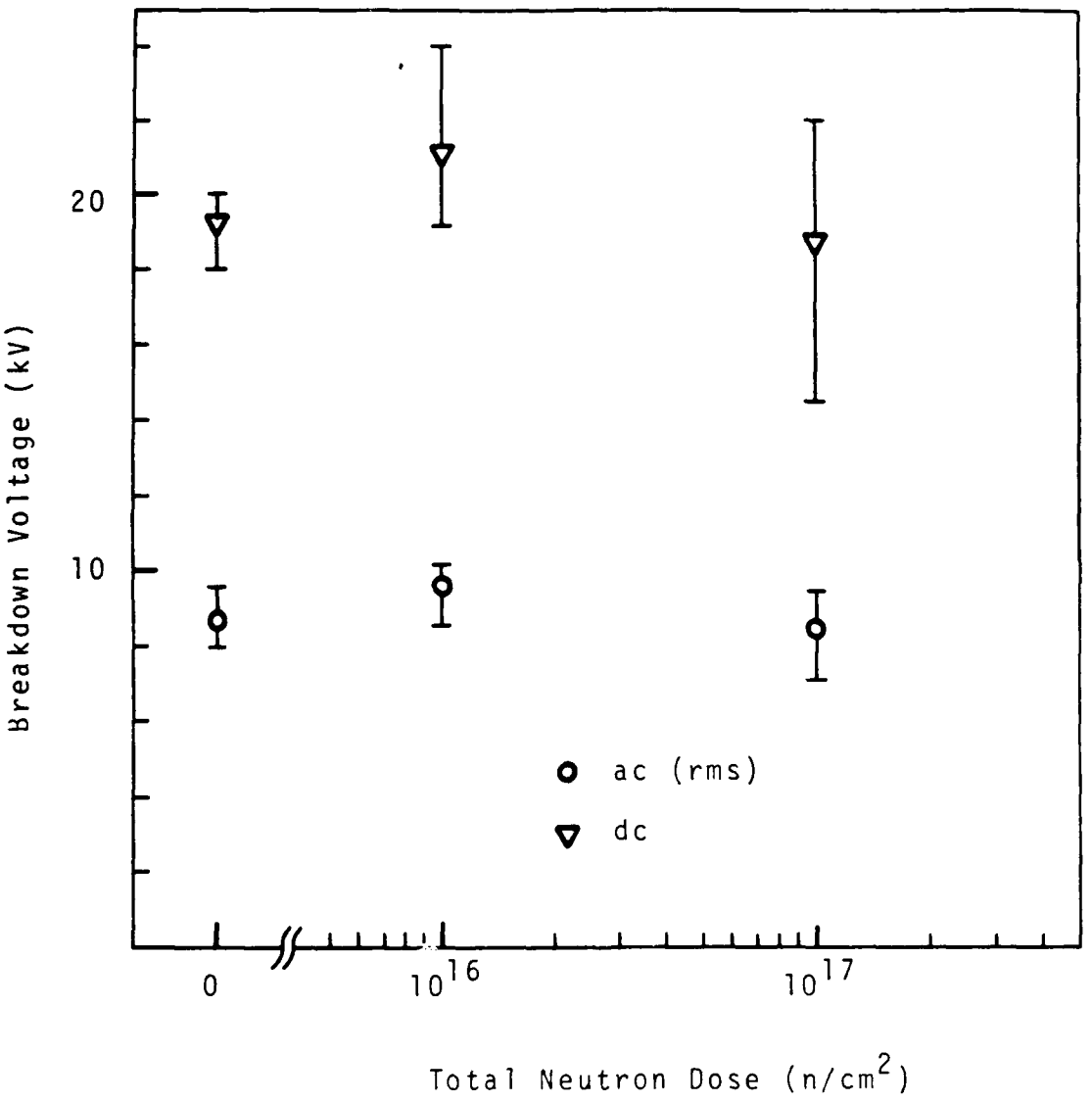

Figure 3.3. ac and dc breakdown voltages versus radiation dose.

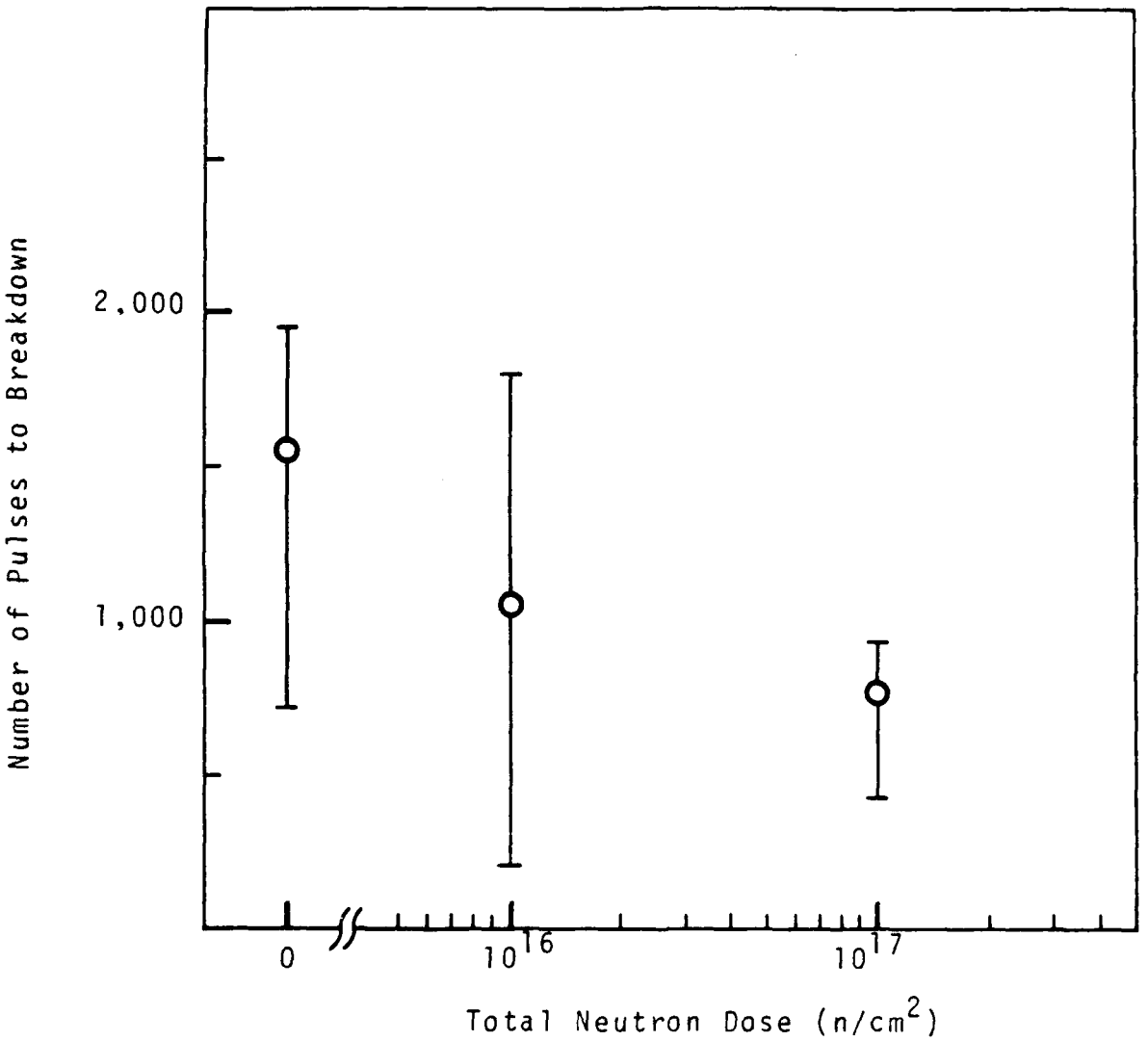

Figure 3.4. Number of pulses to break versus radiation dose.

3. 4 as a function of the neutron dose. The data followed closely the Weibull distribution,[26] therefore, the characteristic life for this distribution is plotted along with 80 percent confidence interval. The line fit to the data points was obtained using the maximum likelihood parameter estimates method for the two- parameter Weibull distribution.[27]

As shown, the lifetime of the irradiated films decreased with increasing dose of absorbed radiation. This is probably due to the increase in the dielectric losses, as was determined earlier, and the consequent increase in the heating of the polymer film, resulting therefore, in premature electro-thermal breakdown [28]. In addition a decrease in the volume resistivity reported later in this chapter has similar effect due to the increase in the joule heating in an applied electric field [9], and subsequent decrease in the pulsed lifetime of the polypropylene film.

The dc volume resistivities of the virgin and irradiated films were measured and the results are given in Table IV. For the virgin sample resistivity of $3.4 \times 10^{18} \Omega.cm$ was obtained. In case of films irradiated for one and ten hours the resistivity dropped down to $2.58 \times 10^{18} \Omega.cm$ for both, which corresponds to 26 percent decrease in the volume resistivity. This decrease can be attributed to the increase in the free carrier concentration

Table IV. Volume resistivities of virgin and neutron irradiated PP films.

Neutron dose (n/cm^2)	**virgin**	**1×10^{16}**	**1×10^{17}**
Volume resistivity ($\times 10^{18}$) (ohm·cm)	3.4	2.58	2.58

produced by the radiation.[5] Moreover, the oxidation mentioned before may have been a factor in the observed phenomenon.

3.3.2. Chemical Changes

There was a noticeable change due to the neutron radiation in the chemical composition of the polypropylene. It is well known that many polymers easily undergo oxidation, which is indicated by the appearance of an absorption band of the C=O group near 1720 cm^{-1}. [29] The oxidation of the polypropylene film under neutron radiation is reflected in Figure 3.5 . Due to the significant increase of the absorption peak around the wavenumber 1720 cm^{-1}, the percentage absorbance for only this peak is given for virgin and irradiated samples. The absorbance increased from 12 percent for the virgin film, to 18 and 20 percent for the films irradiated for one and ten hours, respectively. The C = O stretching vibrations of the carbonyl groups are responsible for the increase in the absorption coefficient at this wavenumber.[30].

3.4. DISCUSSION AND CONCLUSIONS

The results of this work indicate that polypropylene film shows good resistance to neutron radiation up to $3x10^8$rads. Small changes were detected in the electric strength, dielectric

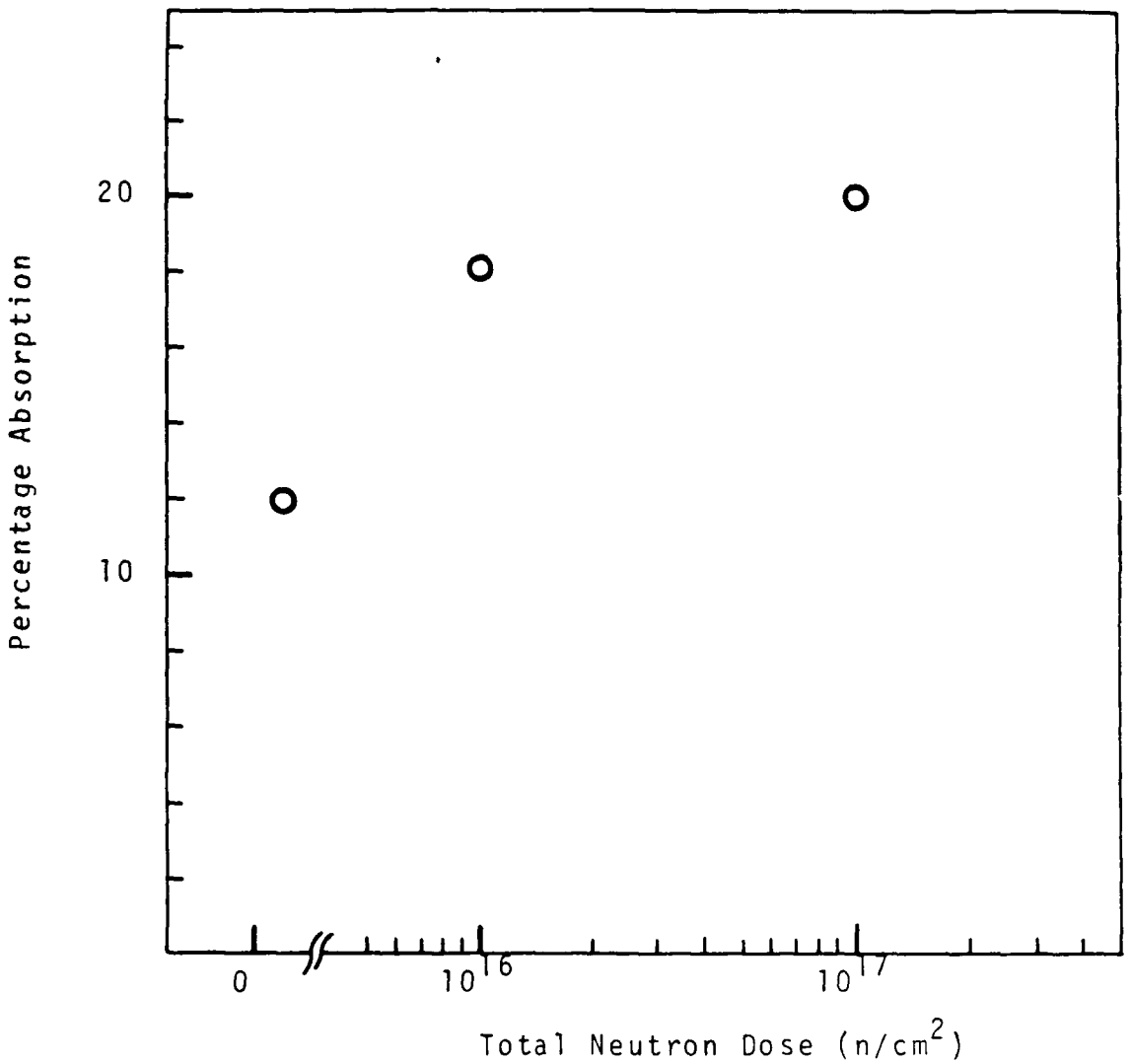

Figure 3.5. % absorbance of the 1720 cm^{-1} peak versus radiation dose.

properties and volume resistivity of the film. The most pronounced were a slight decrease in the breakdown voltage and an increase in the dielectric losses. These changes show that the degradation of the polypropylene film is not very significant for at least $3x10^8$ rads of the total absorbed dose . The oxidation of the polypropylene is believed to be responsible for most of the changes induced in the properties of the film.

The polypropylene film, when used as a dielectric in capacitors located in the containment vessel of the nuclear power plant, can withstand the normal operation dose absorbed over the design life of the plant, which although appreciable, is typically lower than the radiation dose applied to films tested in this work. However, the degradation of the polypropylene film might prove to be too excessive for reliable operation of emergency cooling equipment in case of loss of coolant accident LOCA or rupture of the fuel element container, when the integrated radiation dose can exceed that of tested in this work.

CHAPTER 4

SIMULTANEOUS ELECTRICAL AND THERMAL AGING OF POLYPROPYLENE FILMS

4.1. INTRODUCTION

A large proportion of the failure in electrical components can be attributed to the breakdown of electrical insulation. During the normal operation of an electrical component, the insulation is subjected to multiple stresses such as electrical, thermal and mechanical, which may act together to accelerate degradation and ultimately cause failure of the insulation. Therefore, aging under multifactor stresses is an area that has recently generated a lot of interest. Additionally, simultaneous application of stresses is important as the results are quite different from those obtained when the stresses are applied singly or sequentially [31]. In particular, simultaneous electrical and thermal stresses have been most commonly investigated since the presence of these two stresses is almost unavoidable [32]. A variety of experiments have been performed and models proposed to predict life under both these stresses [33-38].

A large proportion of the aging and lifetime studies has been oriented towards predicting the useful operational lives of cables, which in general bear stresses of the order of 10 V/um only [39]. For the most part, little data is available on the

electrical aging and lifetimes of insulating materials applicable to high voltage capacitors, where the field stresses are amongst the highest of most electrical equipment, around 55 V/um [40,41]. There is now a greater need to operate these capacitors at even higher voltages to yield higher energy densities, particularly for various space power applications, and this has necessitated the use of thin film and other technology to obtain these higher operating stresses in excess of the present upper limits [40]. Very little data is available on the simultaneous high field electrical and thermal aging of capacitor grade films at stresses close to their breakdown strengths.

The present study is aimed at experimentally determining the lifetimes of capacitor-grade biaxially oriented polypropylene (PP) films under simultaneous electrical and thermal stresses, particularly at high ac and dc stresses. Feasibility of applying models to the data obtained and possible mechanisms operating at high electrical stress aging, close to breakdown, are also discussed. These data will be useful in estimating lifetimes and reliability of capacitors operating at the highest possible stresses. This work is also aimed at determining the differences, if any, between ac and dc aging.

4.2. EXPERIMENTAL

The aging test set up is presented in Figure 4. 1. Standard cylindrical brass electrodes 2.54 cm in diameter with rounded edges in accordance with ASTM-D149-81 specifications were used. 25.4 μm-thick biaxially oriented polypropylene film (manufactured by Hercules) was tested with the samples cut to squares of approximately 6 cm. The test cell was filled with transformer oil and heated to the test temperature by means of a hot-plate heater. The whole arrangement was enclosed in a circulation oven for thermal stability over the long duration of the tests. It was found that by this arrangement, a temperature control of ±1 C was obtained. The tests were performed at three temperatures: 23^{o}C, 70^{o}C and 90^{o}C.At each temperature, the ac and dc breakdown voltages (BDV) were determined. Following the BDV test, the lifetimes of samples of the polypropylene film were measured for several voltage levels below the BDV. In each case the voltage was raised at a rate of 500 V/sec. Due to the character of this experiment, namely high field stresses, the data sets are complete i.e. no censoring was required. For each measurement, the estimates of shape and scale parameters were determined by graphical and maximum likelihood methods [27,42,43].

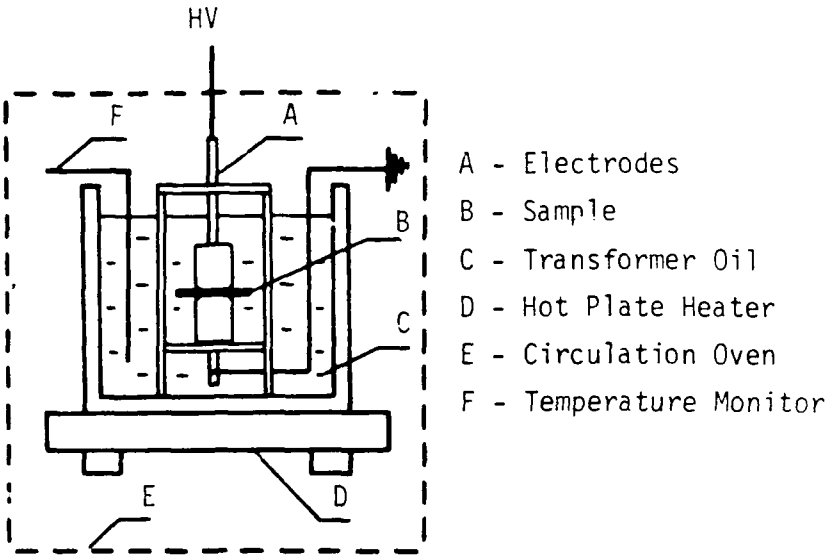

Figure 4.1. Experimental set-up.

4.3. RESULTS

The results of the dc and ac aging experiments are shown in Figures 4.2 to 4.11 and Tables V to VII. Figures 4.2 up to 4.9 are Weibull plots and enable the graphical estimation of the shape and scale parameters. Figures 4.2 and 4.6 represent distribution of breakdown voltages (BDV's) under dc and ac stresses accordingly. The distribution of the failure times at temperatures 23°C, 70°C and 90°C is plotted in Figures 4.3 to 4.5 for dc and in Figures 4.7 to 4.9 for the ac case. In addition, the estimates of the Weibull parameters were determined using maximum likelihood method as shown by Stone and Heeswijk [27]. The scale and shape parameters estimates by the graphical and the maximum likelihood methods are presented in Tables V and VI.

Figures 4.10 and 4.11 are examples of the lifetime data at three temperatures (23°C, 70°C and 90°C), plotted on a log-log scale for dc and ac aging, respectively. In both cases the maximum likelihood estimates of the scale parameter were used.

The scale parameter estimates for the dc lifetimes were also fitted to the models proposed by Ramu [38] and Simoni [36] (Equation 4.4) and Fallou[34](Equation 4.5) and the results are presented in Table VII.

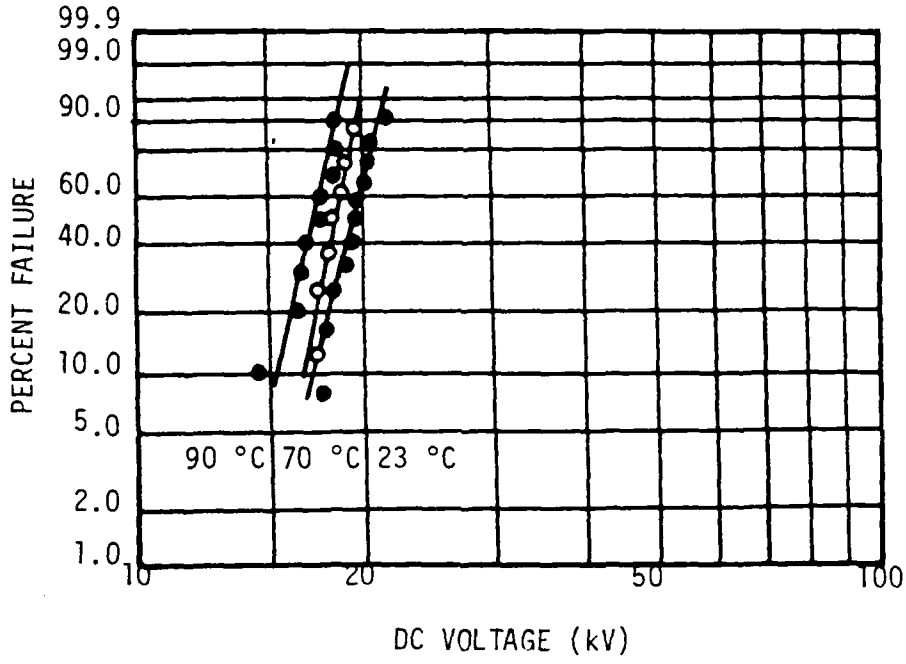

Figure 4.2. Weibull plots for dc breakdown voltages.

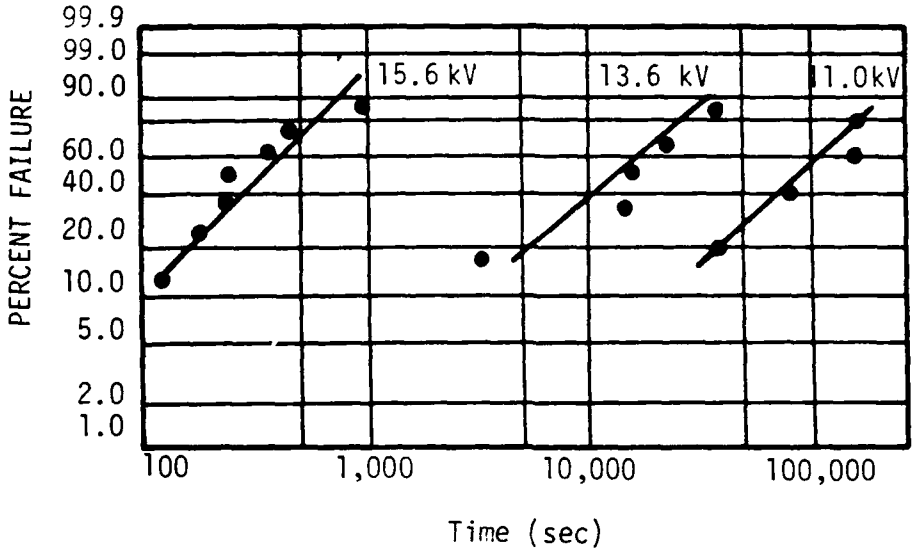

Figure 4.3. Weibull plots for times to breakdown under dc stress at 23 °C.

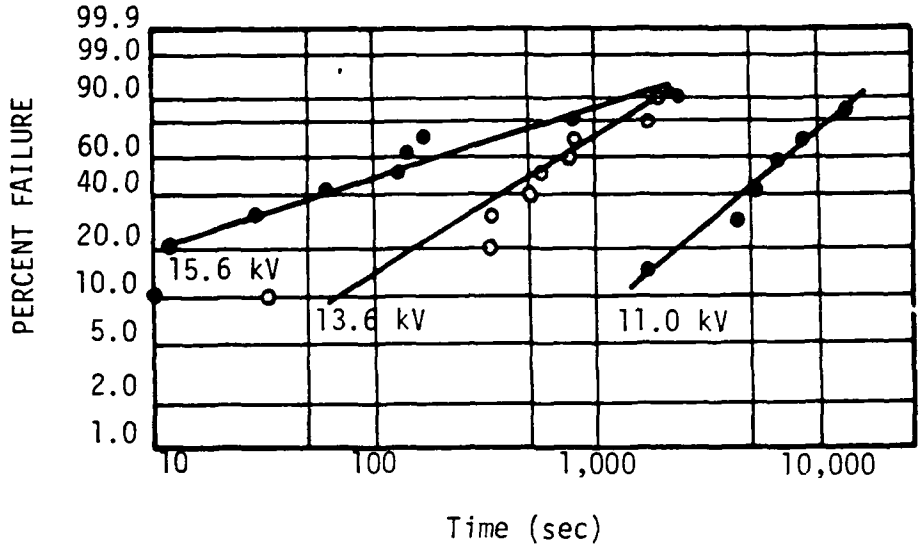

Figure 4.4. Weibull plots for times to breakdown under dc stress at 70 °C.

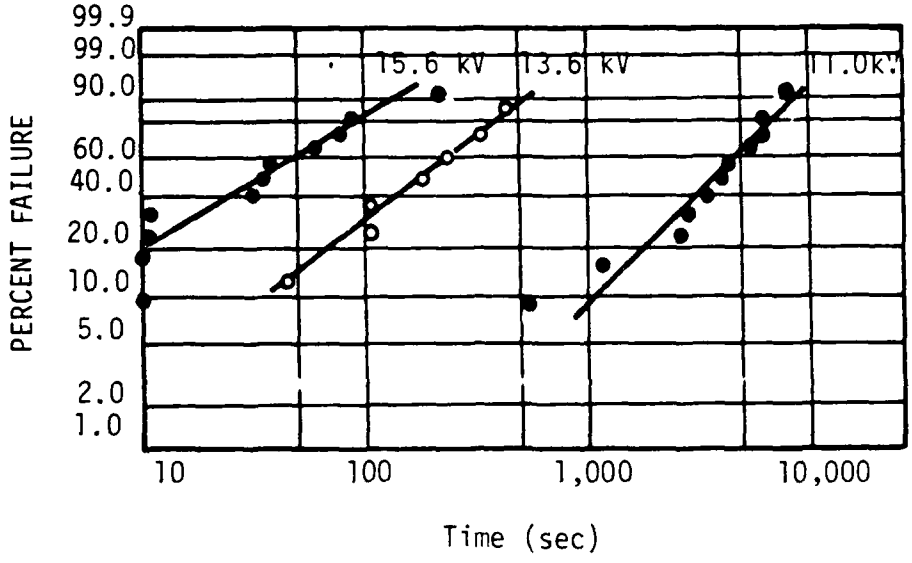

Figure 4.5. Weibull plots for times to breakdown under dc stress at 90 °C.

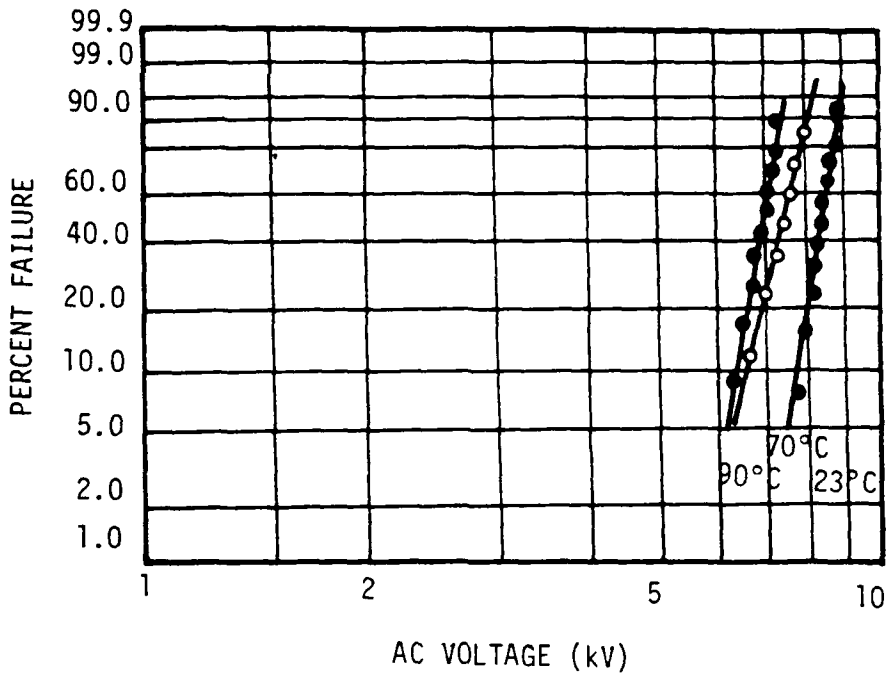

Figure 4.6. Weibull plots for ac breakdown voltages.

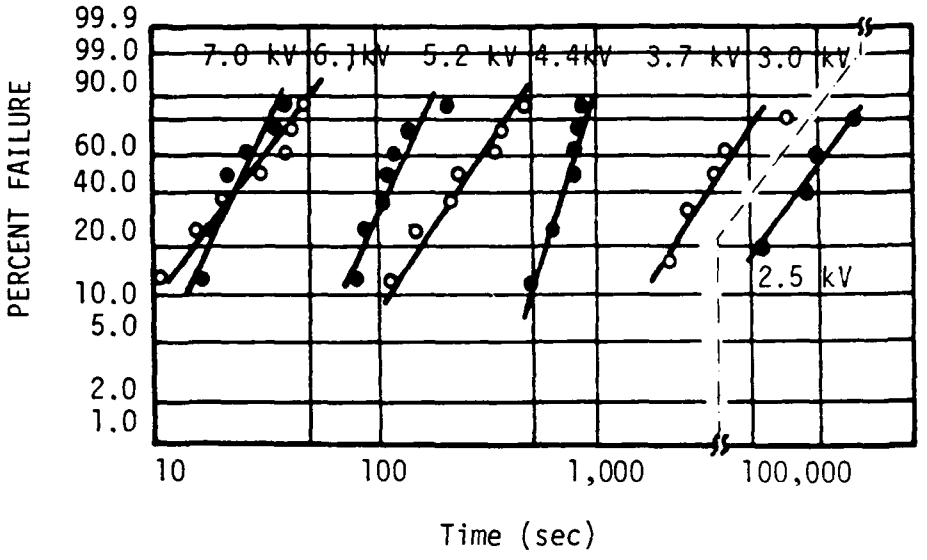

Figure 4.7. Weibull plots for times to breakdown under ac stress at 23 °C.

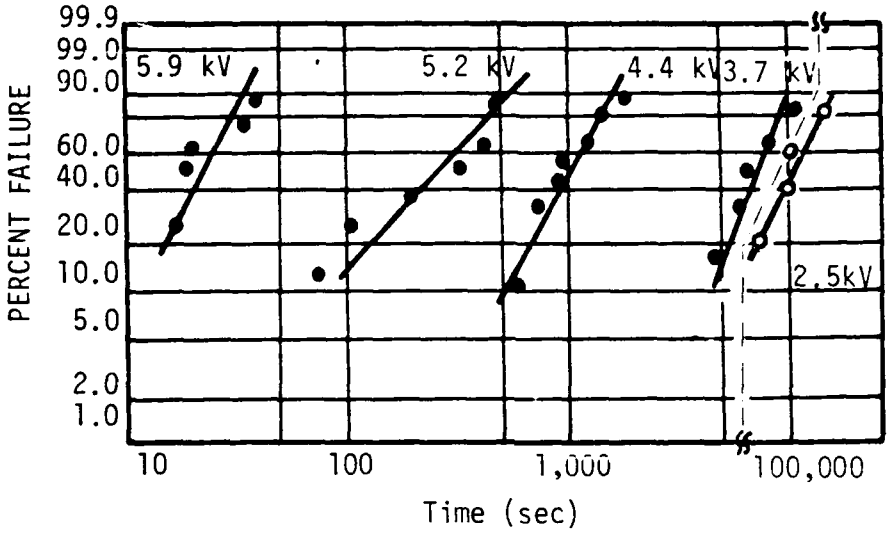

Figure 4.8. Weibull plots for times to breakdown under ac stress at 70 °C.

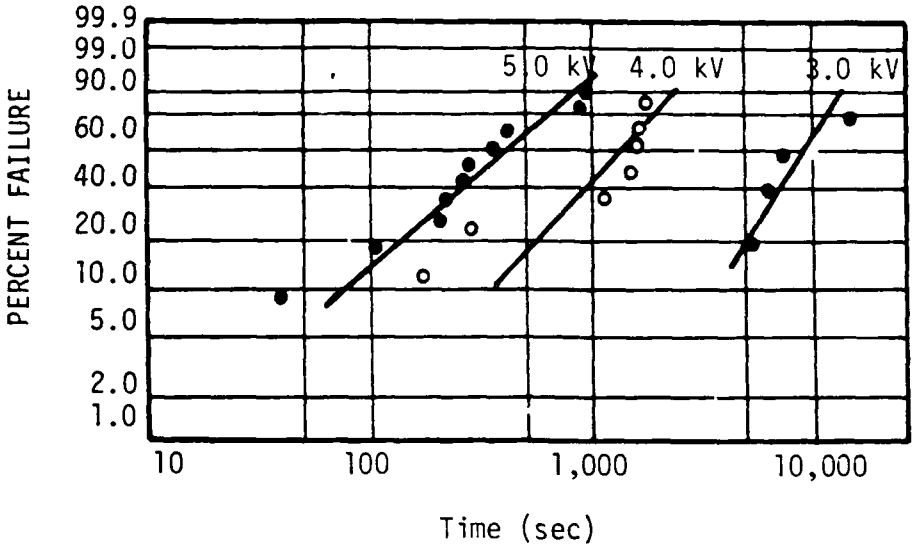

Figure 4.9. Weibull plots for times to breakdown under ac stress at 90 °C.

TABLE V. Graphical and maximum likelihood estimates for lifetimes of PP films under dc stress.

Stresses		Weibull Parameters			
		Graphical		Maximum Likelihood	
Temp. [°C]	Voltage dc [kV]	Scale $x10^3$[sec]	Shape	Scale (90%CI) $x10^3$[sec]	Shape (90%CI)
23 °C	15.6	.41	1.60	.40 (.22,.68)	1.53 (.70,2.19)
	13.6	18.00	1.40	21.70 (9.91,43.96)	1.53 (.55,2.30)
	11.0	130.00	1.32	125.0 (67.9,231.2)	2.44 (.68,3.73)
70 °C	15.6	.28	0.50	.29 (.09,.78)	.64 (.34,.89)
	13.6	.86	0.99	.86 (.46,1.45)	1.22 (.63,1.70)
	11.0	7.80	1.33	7.70 (4.44,12.45)	1.89 (.77,2.76)
90 °C	15.6	.05	0.91	.07 (.04,.10)	1.35 (.75,2.33)
	13.6	.25	1.15	.86 (.50,1.40)	1.63 (.75,2.33)
	11.0	4.80	1.62	7.70 (5.32,10.52)	1.92 (1.06,2.65)

TABLE VI. Graphical and maximum likelihood estimates for lifetimes of PP films under ac stress.

Stresses		Weibull Parameters			
		Graphical		Maximum Likelihood	
Temp. [°C]	Voltage dc [kV]	Scale $x10^3$[sec]	Shape	Scale (90%CI) $x10^3$[sec]	Shape (90%CI)
23 °C	7.0	.03	1.56	.03 (.02,.04)	3.16 (1.45,4.52)
	6.1	.04	1.67	.03 (.02,.05)	2.15 (.99,3.07)
	5.2	.14	2.17	.14 (.10,.18)	3.19 (1.47,4.56)
	4.4	.32	1.84	.31 (.21,.42)	2.49 (1.15,3.56)
	3.7	.85	3.25	.78 (.70,.86)	8.10 (3.72,11.58)
	3.0	3.78	2.74	4.26 (2.53,6.83)	2.29 (.52,3.44)
	2.5	109.08	2.51	109.1 (60.1,193.6)	2.35 (.70,3.53)
70°C	5.9	.03	2.13	.03 (.02,.04)	2.88 (1.32,4.12)
	5.2	.35	1.28	.35 (.22,.53)	1.91 (.88,2.73)
	4.4	1.28	1.91	1.25 (.93,1.61)	2.78 (1.39,3.92)
	3.7	8.25	2.70	8.35 (6.09,11.09)	3.80 (1.37,5.70)
	2.5	117.72	2.23	117.2 (71.2,193.6)	2.98 (.84,4.57)
90 °C	5.0	.44	1.08	.43 (.25,.68)	1.30 (.72,1.79)
	4.0	1.30	1.28	1.32 (.81,2.05)	1.81 (.83,2.59)
	3.0	9.60	2.15	9.76 (5.18,18,48)	2.35 (.67,3.6)

4.4. LIFE MODELS FOR MULTISTRESS AGING

The ultimate goal of lifetime studies is to develop a reliable mathematical model for the aging process relating the test stress to the time to failure. As mentioned before, a number of models for multistress aging have already been proposed [34-38]. Most of these models are extensions of models developed under single stresses, for example, for thermal stress, the Arrhenius relationship [32],

$$L = A \exp[B/T] \tag{4.1}$$

and for the voltage stress, the inverse power model[33,36],

$$L = K V^{-N} \tag{4.2}$$

or the exponential law [35].

$$L = C \exp[-nV] \tag{4.3}$$

The multistress models that have been proposed can be divided into two broad groups --- the ones that have an inverse power law type of dependence for the electrical stress [36,38] and the ones that have an exponential relationship [34,35,37]. The models of both types have been widely used by Simoni [35,36] and by Ramu et al. [37,38] in fitting their data. A general equation for

predicting the lifetimes under simultaneous electrical and thermal stresses with the inverse power term for the electrical aging is given below [36-38].

$$L = k(T)\ \exp(-B\ DT)\ V^{-n(T)} \qquad (4.4)$$

where L is the lifetime, V the voltage stress above Vo, the threshold voltage below which electrical aging is assumed to be negligible, B is the thermal constant, k(T) is temperature dependent constant, given by $k(T) = \exp(k_1 - k_2 DT)$, n(T) is the exponent which is temperature dependent according to the equation $n(T) = n_1 - n_2 DT$ where n_1 and n_2 are constants and DT=1/To-1/T, with T being the absolute temperature.

An example of a model with an exponential dependence for the electric stress is the following [4,7].

$$L = \mathrm{Exp}\ [A(E) + B(E)/T] \qquad (4.5)$$

where $A = A_1 + A_2\ E$ and $B = B_1 + B_2\ E$, A_1, A_2, B_1 and B_2 being constants, and T being the absolute temperature.

4.5. DISCUSSION AND CONCLUSIONS

Figure 4.10 is a plot of the dc voltage vs. the time to breakdown of the test samples at three temperatures --- 23°C, 70°C and 90°C. The inverse power law (Equation 4.4) seems to hold for dc aging, as straight lines result in plotting the data on a log-log graph. The breakdown voltages decreased as the test temperature was raised. Also, the lifetimes of the PP film at 70°C decreased significantly when compared to the life-voltage stress curve at 23°C. A further decrease in the lifetime is observed when the temperature was increased to 90°C. Large power exponent (Table VII) in the lifetime model points to the fact that electrical aging dominates over thermal deterioration under high field stresses. The dc lifetime data was also fitted to the exponential model (Equation 4.5). Table VII also lists the parameters calculated according to this model. However, the inverse power model was found to be a better description of the dependence of lifetimes under dc stresses as a function of the applied voltage even under extremely high electric stresses.

While lifetimes under dc stresses follow the classical model, where the aging processes are accelerated by increasing the temperature, behavior under ac stresses follows an anomalous pattern (Figure 4.11). Three regions were identified in the ac aging case--- namely high, intermediate and low electric field

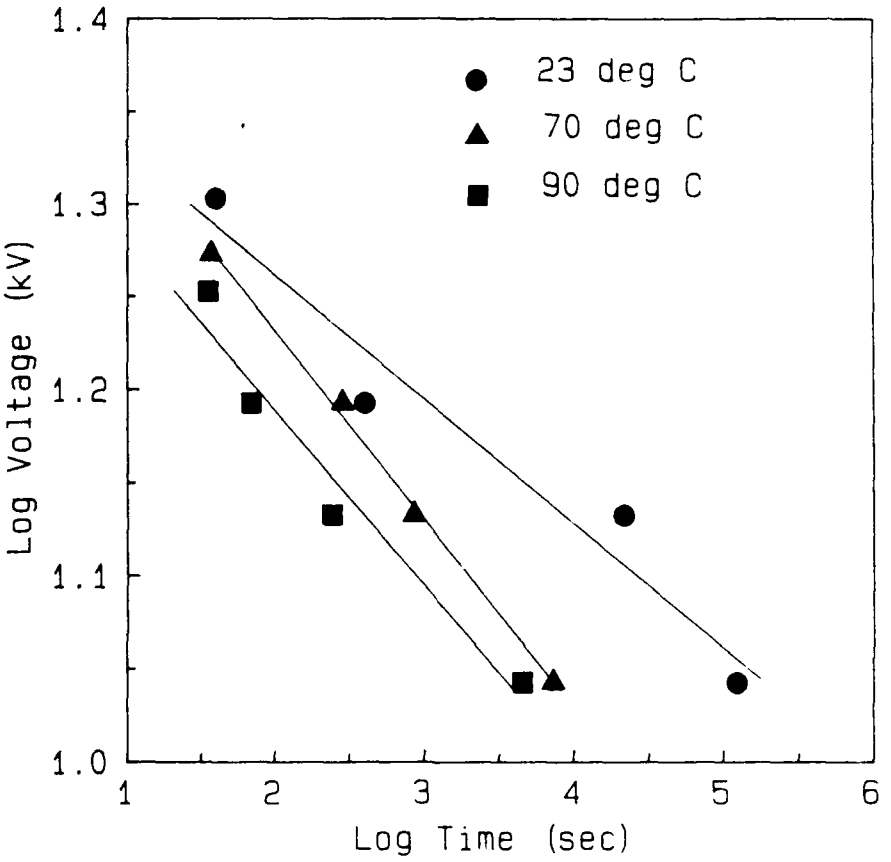

Figure 4.10. Lifetimes under dc stress.

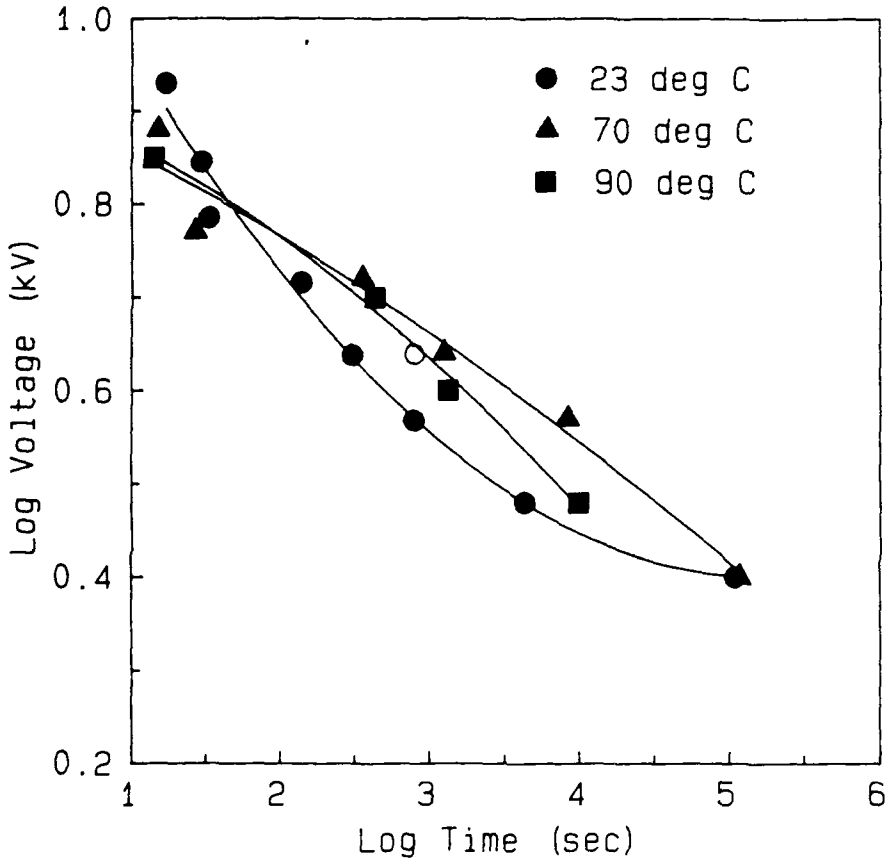

Figure 4.11. Lifetimes under ac stress.

(O —— impregnated sample)

Table VII. Calculated parameters of life equations for simultaneous electrical (dc) and thermal stresses.

Models	Parameters of Life Equations			
Inverse Power (Equation 4)	n_1	n_2	k_1	$k_2 + B$
	14.11	4.88×10^3	46.22	1.60×10^4
Exponential (Equation 5)	A_1	A_2	B_1	B_2
	-18.95	0.85	12.99×10^3	-5.99×10^2

regions. It is speculated that these regions arise because of the different mechanisms that operate at different electric stresses and temperatures. The anomaly is concentrated in the intermediate or transitional region where the lifetimes at 23°C are the longest and those at 70°C the shortest with the 90°C curve falling somewhere in between. Probably the biggest factor in the existence of the transition region in moderate electric field is the progressive impregnation of the dielectric films with the transformer oil competing with the thermal aging of the films. The efficiency of impregnation can be expected to increase with increasing temperature [44]. If this is the case, the 70°C lifetime curve represents the optimal impregnation temperature out of the three used in the experiments. The 23°C curve would represent incomplete impregnation and the 90°C curve would represent the deterioration due to thermal aging counteracting the effects of impregnation. In the first region of high electric stress, electromechanical breakdown may have been the predominant factor in the breakdown of the insulating film. Lower elastic moduli at higher temperatures would lead to lower breakdown voltages and lifetimes [19,45]. When the electric stress is decreased, partial discharges and thermal aging processes become responsible for the failure. It is in this intermediate region that impregnation can play a significant role, as impregnation helps to diminish the partial discharges. This effect is especially prevalent under ac

stresses where partial discharges occur to a greater degree and at a higher frequency than under dc stresses [46]. At low electric fields, the impregnation process is essentially complete and the chemical processes with rates dependent mostly on the temperature play the major role in the failure mechanisms. Impregnation was not expected to be a factor in the case of dc because partial discharges are not identified as a significant degradating factor [46]. This was confirmed by the absence of any transition region. The ac life dependence on the voltage stresses was found not to be amenable to any empirical analysis.

To confirm the influence of impregnation on lifetimes of the polypropylene film in the intermediate region, the times to breakdown of impregnated samples were also determined. Impregnation temperature was chosen to be 70°C, which was established to be optimal out of three temperatures applied in aging studies, and the sample was then aged under 23°C. The results confirmed the trend expected, i.e. the lifetimes of these impregnated samples were considerably higher than the ones not impregnated and aged under room temperature (see Fig. 4.11).

The present study shows that there is a need to give physical foundation to models that seek to predict the lifetime of an insulator under simultaneous electrical and thermal stresses. If this can be achieved, the variation of the parameters used in

these models with electric field and temperature can be predicted without recourse to a totally empirical approach.

CHAPTER 5

CONCLUSIONS

5.1. SUMMARY OF PRESENT WORK

A biaxially-oriented, 25.4 μm-thick, capacitor-grade polypropylene film was subjected to electron and neutron/gamma radiation in order to assess its performance under different types of ionizing radiation. A number of electrical and mechanical properties including the breakdown characteristics, the dielectric constant, the dissipation factor and the tensile properties were monitored to determine the degree of degradation suffered by the film. The results of these studies, described in Chapters 2 and 3, can be summarized as follows:

- Radiation-induced changes in the polypropylene film whether from electron or neutron/gamma radiation, were a result of cross linking and/or chain scission.

- Impregnation with MIPB seemed to have a stabilizing effect on the degradation of some of the properties. For example, the dissipation factor of the impregnated films exhibited a downward trend with increasing electron radiation dose.

- o The life of polypropylene films under electron radiation can be increased up to 10^8 cumulative rads if impregnated with a suitable dielectric fluid such as MIPB. Dry polypropylene films can tolerate up to 3×10^8 rads of reactor radiation at a fluence rate of 2.6×10^{12} $n_o/cm^2.s$ accompanied by gamma radiation at a level of 10^7rads/hour

Lifetimes of polypropylene films under simultaneous electrical and thermal stresses were determined with a view towards completing the multifactor stress (E/T/R) aging matrix. The films were subjected to both dc and ac electric fields while immersed in transformer oil at 23°C, 70°C and 90°C. Unlike most previous studies the electric fields employed were close to the breakdown strengths of the films in order that realistic extrapolations could be made towards high energy density capacitor operating voltages. The following are the more important points that emerged from this study:

- o Partial discharges are probably the most important aging mechanism under ac conditions. Therefore impregnation of the films with a dielectric fluid would effectively increase its lifetime.

- o Under dc conditions, the lifetimes of the PP films

obeyed an exponential or power-law model with an Arrhenius type of relationship for the temperature dependence.

- The ac lifetime data could not be fitted to any model for the entire range of voltage because of the different mechanisms that are thought to be prevailing at the different electric field regions and also because of the progressive impregnation of the films with transformer oil.

5.2. FUTURE WORK

Work is currently underway to complete the multifactor stress (E/T/R) aging matrix. This work is expected to consist of two main phases. The first phase will be the gathering of the lifetime and/or performance degradation data at the SUNY at Buffalo research reactor facilities. These studies will cover various combinations of electrical, thermal and radiation stresses on the polypropylene film. The second phase will take the form of developing models to enable the extrapolation to normal operating conditions of the capacitors.

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APPENDIX A

EFFECT OF HIGH ENERGY ELECTRON RADIATION ON MIPB-IMPREGNATED POLYPROPYLENE FILM

by

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ABSTRACT

This work examines the effect of impregnating capacitor-grade polypropylene film with an aromatic hydrocarbon fluid with a view towards assessing the response under radiation of the relevant properties of the film. Monoisopropyl biphenyl (MIPB)-impregnated polypropylene film was exposed in air to 1 MeV electrons to different doses and the post-radiation effects on the electrical, mechanical, morphological and chemical properties were evaluated. The results obtained indicate that the oil-impregnated films remain relatively stable even at the highest dose of 10^8 rads. This is in contrast to the results reported earlier for dry polypropylene films[1,2] which underwent degradation primarily due to chain scission and oxidation. In the present instance, it is believed that impregnation of the film has inhibited the diffusion of oxygen and consequently favored crosslinking. The chemical characterizations as well as the changes in some of the measured properties of the film seem to corroborate this belief.

INTRODUCTION

In high voltage space power systems, there is a demand for increasing the energy densities as well as the power levels. In addition, components with longer lifetimes and better reliability are required. Insulating materials which are commonly employed within these systems as dielectrics and supporting structures, are often the weak links as their degradation and breakdown precipitate the failure of the equipment. This degradation can be accelerated by the exposure to radiation in environments such as space and nuclear reactors. These materials, therefore, play a key role in the reliable design and performance of these systems.

Polymers nowadays constitute the largest class of insulating materials in use because of their excellent electrical and mechanical properties.[3] In addition, their relatively low densities make them good candidates for applications where weight and volume limitations are important. Polypropylene is one such polymer which finds major application in high energy density capacitors.[4] In high voltage applications, the dielectric film is often impregnated with a dielectric fluid to improve its reliability and life by way of suppressing discharges within voids and gas pockets.[5] The fluid also helps in transferring away the heat that builds up during the operation of a capacitor.[4] Monoisopropyl biphenyl (MIPB) is an aromatic hydrocarbon liquid that possesses good dielectric properties, low viscosity and high gas absorption ability[5] and is commonly used as an impregnant for high voltage capacitors. In addition, this liquid is known to exhibit excellent radiation stability.[6]

To investigate the influence of radiation on a typical dielectric system used in high voltage and energy storage capacitors, MIPB-impregnated polypropylene film was exposed to high energy ionizing radiation. Electron radiation, which has been reported as influencing the performance of many space vehicles and on-board electronic components,[7,8] was chosen in this work. The energy of the striking electron beam was 1 MeV because spontaneous internal breakdowns in many spacecraft dielectrics have been reported to occur at about this energy level.[9] In addition, electrons having energy levels of 1 MeV and higher are encountered in space,[9,10] either directly or through secondary emissions. The post-radiation effects on the electrical, mechanical, morphological and chemical properties were investigated and are reported as functions of the total absorbed dose. The electrical properties included the dielectric constant, dissipation factor, the ac (60 Hz) and dc breakdown voltages. The mechanical properties investigated comprised the Young's modulus, elongation-at-break, tensile strength, storage modulus and mechanical loss. The morphological and chemical characterizations included optical and scanning electron microscopy and infrared spectroscopy.

EXPERIMENTAL

A 25.4 μm-thick biaxially-oriented polypropylene film, manufactured by Hercules, Inc., was used as the solid dielectric. The film was impregnated with MIPB, an aromatic hydrocarbon fluid. Some of the properties of interest of the film and the impregnant are listed in Tables I[11] and II,[5,12] respectively.

The oil-impregnation of the films was done as follows: samples were first dried in a vacuum oven at 10^{-2} Torr at 50 °C for 6 hours. The liquid impregnant was then bled into the oven till the samples were completely immersed in the fluid. The samples were then allowed to soak in MIPB for 16 hours at 50 °C under vacuum. At the end of this period, the samples were allowed to cool slowly to room temperature and dry nitrogen was bled into the oven to bring the pressure up to atmospheric. The impregnated samples were then stored in a nitrogen atmosphere.

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Table I. Properties of the polypropylene film.[11]

Dielectric constant, 24 °C, 60 Hz & 1 kHz	2.3
Dissipation factor, 24 °C, 60 Hz & 1 kHz	0.0002
Dielectric strength, 24 °C, 60 Hz (V/μm)	315
Specific gravity	0.902
Weight-percent crystallinity	56
Tensile modulus (N/mm^2)	2,420
Elongation (%)	70-100
Tensile strength (N/mm^2)	205
Melting point (°C)	167
Glass transition temperature (°C)	-18

Table II. Properties of MIPB.[5,12]

Classification	80% aromatic hydrocarbon
Dielectric constant, 25 °C, 60 Hz	2.8
Dielectric strength, 25 mm gap (V/μm)	2.8
Specific gravity	1.0
Viscosity, 38 °C (cs)	6.1
Solubility in water	Insoluble
Boiling point (°C)	270
Flash point (°C)	155
Pour point (°C)	-55

While the radiation procedure of the films as well as the techniques adopted in performing the electrical, mechanical, morphological and chemical measurements have been given in detail in the previous papers,[1,2,13] it is worth enumerating the following important points of the experimental procedure:

- The films were exposed in air to 1 MeV electrons at a rate of 10^6 rads/min. Samples with total radiation doses of 10^5 rads, 10^6 rads, 10^7 rads and 10^8 rads were obtained. During each irradiation run, the samples were stacked in a tray with the excess oil being removed in order to ensure uniform exposure and to prevent the attenuation of the electron beam.

- For the breakdown voltage measurements, two cylindrical brass electrodes of 2.54 cm diameter were used and the rate of voltage rise was 500 V/s. To prevent surface flashover, the breakdown tests were conducted in a test cell filled with transformer oil.

- For the tensile tests, strips having the dimensions 5 cm x 6 cm were extended at a rate of 20 mm/min. The dynamic mechanical properties were determined in a temperature range from 12 °C to 120 °C. These measurements were made at a frequency of 110 Hz and the rate of increase of the temperature was 3 °C/min.

The data obtained on the electrical and tensile properties represent the average values of at least seven measurements. The error bars given indicate the maximum and minimum values.

RESULTS AND DISCUSSION

Electrical Properties

Dielectric characterization of the films was carried out in terms of changes in their dielectric constant and dissipation factor with the radiation absorbed dose at six frequencies, ranging from 50 Hz to 10 kHz.

The dielectric constant of the impregnated film did not display any dependence on the frequency. Therefore, only the data obtained at 50 Hz is presented in Figure 1. It can be clearly seen that the film suffered a slight reduction in its dielectric constant as the absorbed dose was increased. At 10^8 rads, for example, this property had decreased by 15% of its unirradiated value. Such a reduction of the dielectric constant with increasing dose may point towards the crosslinking of the polymer.

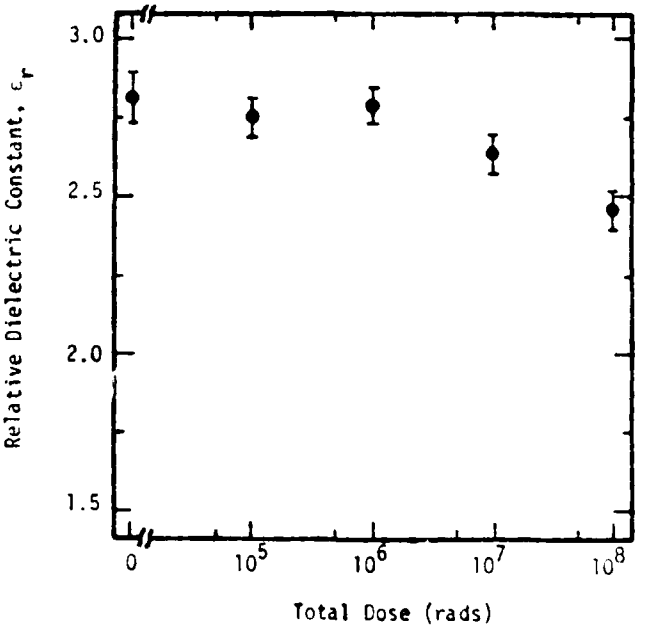

Figure 1. Relative dielectric constant at 50 Hz versus total radiation dose.

Unlike the dielectric constant, the dissipation factor of the film exhibited a significant dependence on the frequency of measurement. The values of this property at four frequencies are plotted against the absorbed

dose in Figure 2. While there was a significant decrease in the dissipation factor with increasing radiation dose at low frequencies, the effect is less dramatic at high frequencies. This seems to indicate that crosslinking of the polymer has an inhibitory effect only on the motions of the larger molecular segments which are responsible for the losses at lower frequencies. The orientational motions of the smaller molecular segments, which are persumably responsible for the losses at higher frequencies, are not affected by the large scale interlinking of the molecules.

A comparison of the ac (peak) and the dc breakdown voltages are shown in Figure 3. While the dc breakdown voltage dropped initially with the first exposure to radiation, it remained unaffected by further radiation. The ac breakdown voltage did not exhibit any significant changes with irradiation. Because of the large spread in the data of both the ac and dc breakdown voltages, it is not clear whether impregnation of the film has had any influence on the impact of radiation with regard to the breakdown behavior.

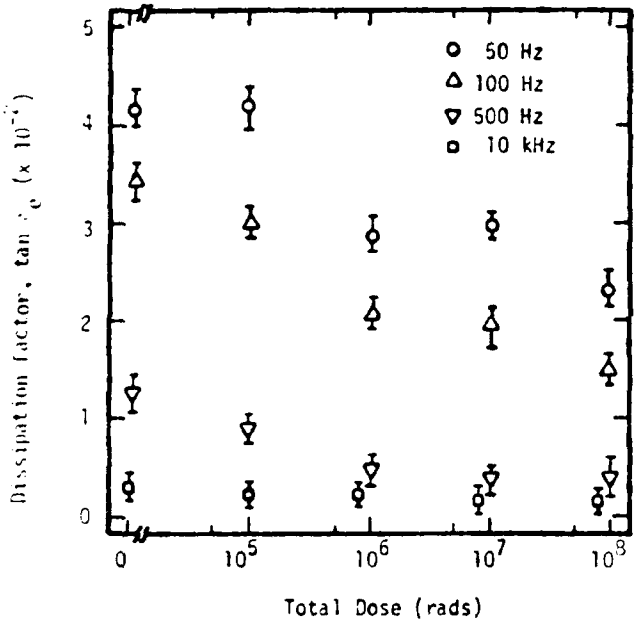

Figure 2. Dissipation factor versus total radiation dose.

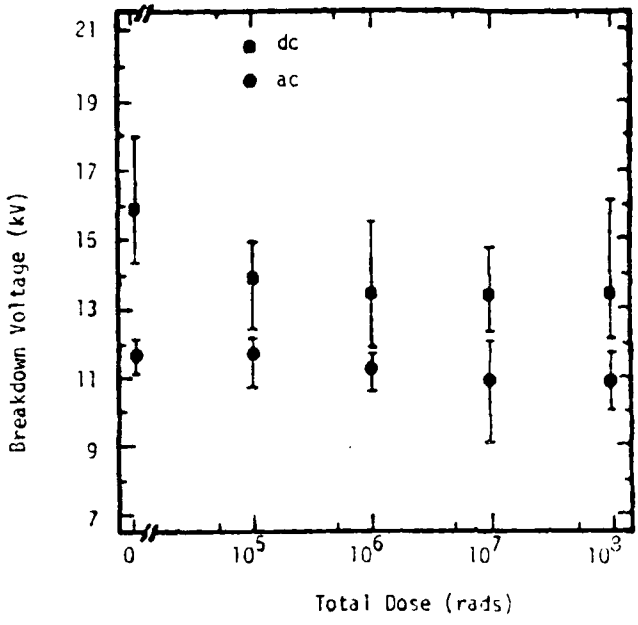

Figure 3. Breakdown voltage versus total radiation dose.

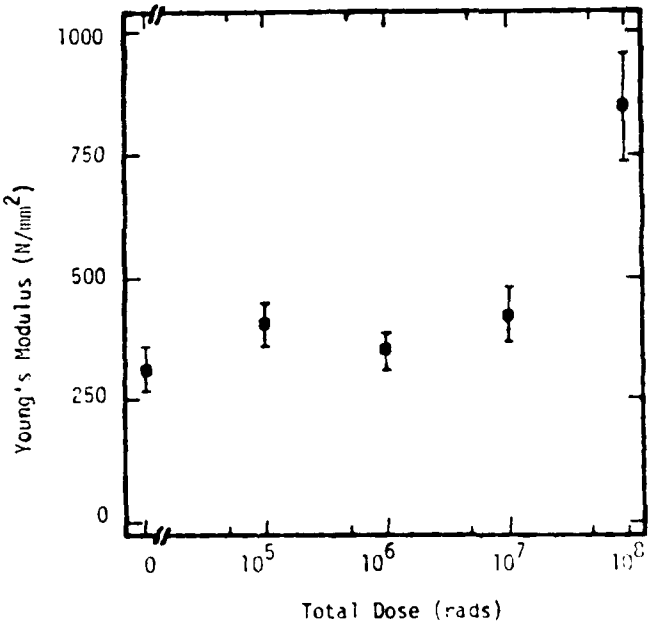

Figure 4. Young's modulus versus total radiation dose.

Mechanical Properties

The variation in the Young's modulus of the impregnated film as a function of radiation absorbed dose is shown in Figure 4. It is seen that the Young's modulus exhibits a gradual increase with increasing dose, reaching at 10^8 rad dose level a value which was more than double of what it was before exposure to radiation. As crosslinking is known, in general, to increase the Young's modulus of polymers,[14] the data obtained support the content on that the impregnated films underwent crosslinking.

Figure 5 illustrates the changes in the elongation-at-break of the film as a function of the radiation absorbed dose. After showing little change with increases in the dose up to 10^6 rads, the elongation-at-break decreased with further increases in the dose. At 10^8 rads, this property lost about 30% of its unirradiated value. This decrease in the material's ability to stretch at any given stress may once again be symptomatic of the crosslinked structure existing in the irradiated samples of the polypropylene film.

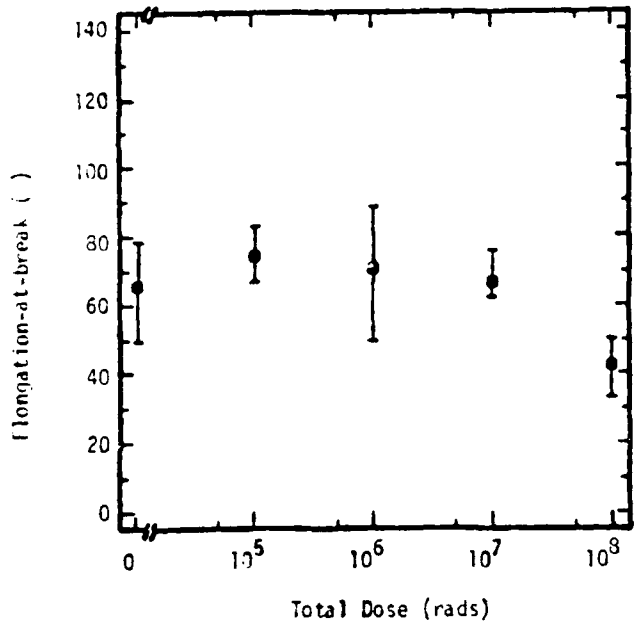

Figure 5. Elongation-at-break versus total radiation dose.

Unlike the Young's modulus and the elongation-at-break, the tensile strength of the film did not show any variation with radiation dose (Figure 6). Hegazy et al.[15] have reported similar trends for the tensile strength as well as for the elongation-at-break of isotactic polypropylene when exposed to gamma radiation under vacuum.

The dynamic mechanical properties of impregnated films irradiated to different total doses are shown in Figures 7 and 8. The temperature range (12 °C to 120 °C) in which these measurements were made corresponds to the region between the glass transition (beta) and the pre-melting relaxation (alpha) of isotactic polypropylene. The data show that radiation has not caused any significant effect on either the storage modulus (Figure 7) or the mechanical loss (Figure 8) until a dose of 10^8 rads is reached. These changes, which appear primarily above 40 °C, consist in a decrease in the modulus

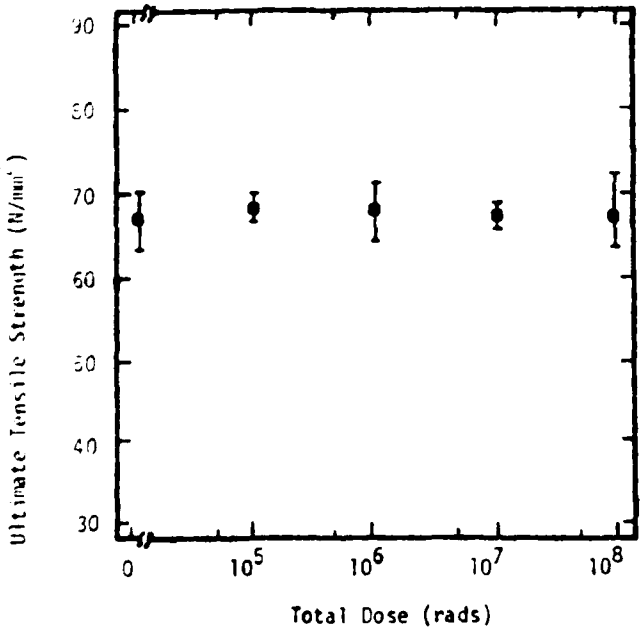

Figure 6. Ultimate tensile strength versus total radiation dose.

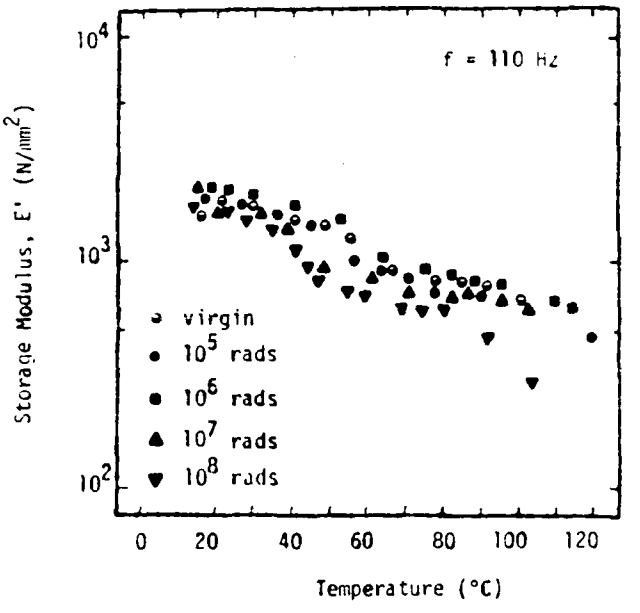

Figure 7. Variation in the storage modulus with temperature for different total radiation doses.

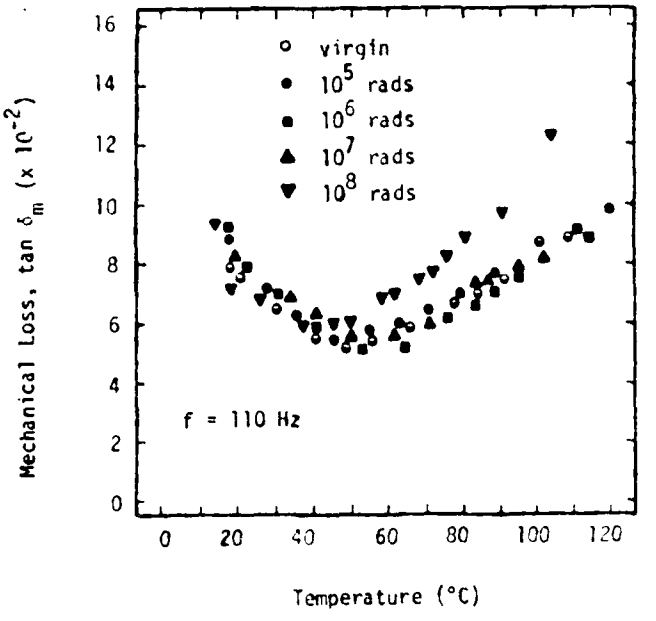

Figure 8. Variation in the mechanical loss tangent with temperature for different total radiation doses.

and an increase in the loss of the 10^8 rad sample. The precise nature of the alpha relaxation, in which region most of the changes are observed in the present study, is not unambiguously known.[16] It is possible that the destruction of the crystalline order by crosslinking is responsible for the increase in the loss or the equivalent decrease in the storage modulus of the 10^8 rad sample. Until the dynamic mechanical measurements are extended to include a wider range of temperatures (particularly below 0 °C), the data pertaining to these properties would remain inconclusive.

Morphological and Chemical Characterization

While the dry film exhibited changes in its surface morphology, as previously reported,[2] especially at 10^8 rads, the optical and scanning electron microscopic studies performed on the impregnated film did not reveal any differences between the unirradiated and irradiated samples.

Similarly, chemical characterization of the irradiated impregnated films obtained through the use of infrared spectroscopy did not show any radiation-induced changes. This is in contrast to the results for the dry film which, when irradiated to 10^8 rads, underwent oxidation as evidenced by the appearance of peaks characteristic of the carbonyl group.[2] It is believed that the barrier presented by the impregnating fluid to the free diffusion of oxygen during radiation has been instrumental in preventing oxidative degradiation. This would, in turn, mean that chain scission is suppressed and crosslinking is favored.[17] These results taken together with the increase in the Young's modulus with radiation support the earlier belief that changes that occurred in some of the film properties are attributable to crosslinking.

CONCLUSIONS

The present work indicates that polypropylene, widely employed in cables and capacitors, can be reliably used in radiation environments when they are impregnated with a suitable dielectric fluid, such as MIPB. When exposed to high energy electron radiation with doses as high as 10^8 rads, the impregnated films showed good tolerance and stability in terms of their electrical and mechanical properties. This stability to radiation comes about due to the presence of the impregnant which seemed to have encouraged crosslinking in preference to chain scission and oxidation degradation.

ACKNOWLEDGMENTS

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APPENDIX B

CALCULATION OF ABSORBED ENERGY

Energy absorbed is a ratio of the energy imparted by the ionizing radiation to the matter in a volume element to the mass of the matter in that volume element. The absorption of energy in a material by an indirectly ionizing radiation such as neutron radiation is a two step process. In the first step, the indirectly ionizing radiation interacts with the matter to produce charged particles, becoming then directly ionizing radiation. In the second step the charged particles impart energy to the medium. The energy transferred in the first step is called kerma, while the absorbed dose represents the results of the second step.

The kerma in the ambient medium and the absorbed dose are related. Under the conditions of approximate charged particle equilibrium, the kerma in the ambient medium and the absorbed dose have nearly equal magnitudes[47]. Since there are tables published [48] of kerma versus energy of impinging neutrons, and since with a good approximation the absorbed energy can be compared with the kerma, the latter was calculated for the energy spectrum of neutron radiation to which polypropylene film samples in this study were exposed. The energy spread of neutrons shown in Figure B1 is sufficiently large, so that an appropriate

average over the values in the tables can be used.

For monoenergetic neutrons the kerma can be calculated in the following way:

$$K = \Phi(\mu_k/\rho) \qquad \text{(B.1)}$$

where, K - kerma

Φ - fluence of the indirectly ionizing particles

(μ_k/ρ) - mass energy transfer coefficient

E - energy of the neutrons

For a spectrum of neutrons, where $\Phi(E)$ is the fluence of neutrons with energies between 0 and E, kerma can be expressed as follows:

$$K = \int \frac{d\Phi(E)}{dE} (\mu_k/\rho)_E \, dE$$

For an approximate calculation of kerma the following equation can be used:

$$K = \sum \Delta\Phi_L \left[\overline{(\mu_k/\rho)E}\right]_L$$

where, L - the energy interval

$\Delta\Phi_L$ - number of neutrons within the energy interval L

$\left[\overline{(\mu_k/\rho)}\right]_L$ - arithmetic average of tabulated kerma per unit

fluence in the energy interval L

The percentage weight of carbon and hydrogen in the propylene monomer is 85.63% and 14.37%, respectively. From the kerma tabulated for carbon and hydrogen, the kerma for polypropylene can be calculated as linear weight ratio of both. For the neutron energy spectrum given in Figure B.1, the kerma calculated from Equation B.3, is equal to 8.1×10^5 erg/g*s, which converts

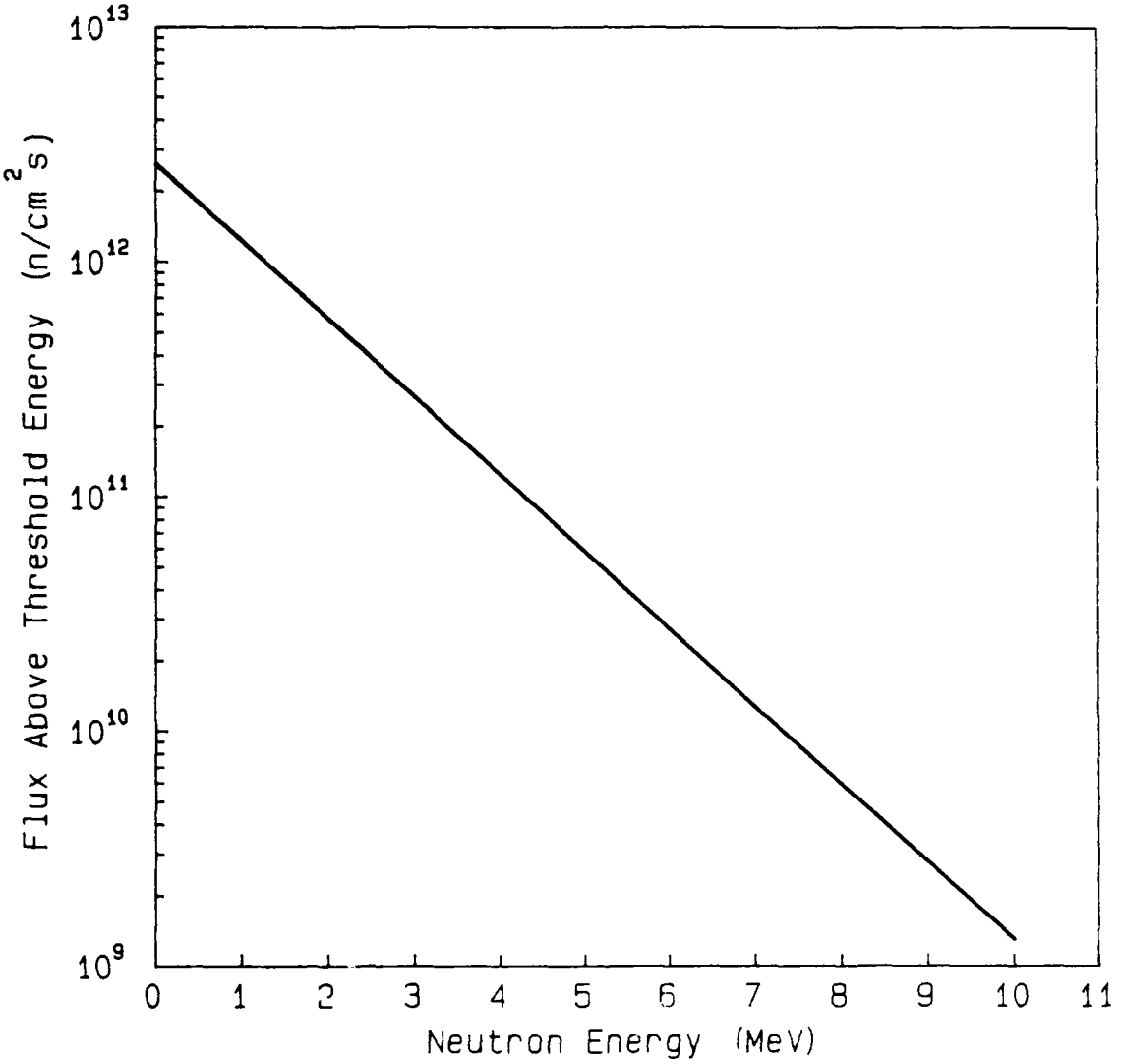

Figure B-1. Neutron flux energy spectrum in the pneumatic conveyer of the thermal nuclear reactor.

to $8.1x10^3$ rad/s. For a total neutron flux equal to $2.6x10^{12} n/cm^2*s$, 1 rad of absorbed dose by polypropylene film corresponds to $3.2x10^8$ n/cm^2 of the neutron fluence. This number is in good agreement with the conversion given elsewhere,[48] where 1 rad is reported as equal to $4x10^8$ n/cm^2. Therefore, the total neutron dose of $1x10^{16}$ and $1x10^{17}$ n/cm^2 is equivalent to $2.9x10^7$ and $2.9x10^8$ rads of energy absorbed, respectively. It is to be mentioned, that the neutron radiation is accompanied by the gamma radiation at the level of 10^7 rads/hour and that the gamma-rays contribute significantly to the matter-radiation interactions.